ORIGINAL PATENT APPLICATION BASED ON:

Docket: **85588RLO**

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Quang Phan

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ELECTROLUMINESCENT DEVICES HAVING CONJUGATED ARYLAMINE POLYMERS

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ELECTROLUMINESCENT DEVICES HAVING CONJUGATED ARYLAMINE POLYMERS

FIELD OF THE INVENTION

The present invention relates to electroluminescent (EL) devices having conjugated arylamine polymers.

BACKGROUND OF THE INVENTION

Electroluminescent (EL) devices such as light emitting diode (LED) are opto-electronic devices which radiate light on the application of an electrical field. Organic materials including both polymers and small molecules have been used to fabricate LEDs. LEDs fabricated from these materials offer several advantages over other technologies, such as simpler manufacturing, low operating voltages, and the possibility of producing large area and full-color displays. Organic polymers generally offer significant processing advantages over small molecules especially for large area EL display because polymer films can be easily produced by casting from solutions.

Conjugated polymers such as poly(phenylvinylene) (PPV) were first introduced as EL materials by Burroughes et al in 1990 (Burroughes, J. H. *Nature* 1990, *347*, 539-41). Other conjugated polymers include poly(fluorene) (PF), poly(p-phenylene) (PPP), and poly(thiophene). Due to the rigidity of the polymer backbone, the polymers are insoluble without introducing the flexible side chains. Linear or branched alkyl or alkoxy are the most commonly utilized solublizing groups. PPVs and PFs and their derivatives are among the most studied conjugated polymers because of their great potential applications in various areas including LED, photodiodes, organic transistors, and solid state laser materials. Electron donor such as alkoxy substitued PPVs show higher efficiencies than unsubstituted ones in LED applications. Other substituents have been rarely investigated. Amine groups are stronger electron donors than alkoxy groups, and amino-substituted PPVs have also been prepared to investigate the effect of amino groups on the LED efficiencies. However, only dialkylamines have been incorporated into PPV as substitutents (Stenger-Smith, J. D. et al *Macromolecules* 1998, *31*, 7566-7569). It is known that dialkylamino groups are susceptible to oxidation.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide polymeric luminescent materials useful for EL devices.

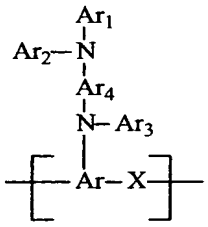
It is a further object of the present invention to provide various energy bandgap luminescent polymers which emit broad range of color.

It is another object of the present invention to provide low ionization potential polymers useful as hole injection materials in EL devices.

These objects are achieved in an electroluminescent device, comprising:

a) a spaced-apart anode and cathode; and

b) an organic layer disposed between the spaced-apart anode and cathode and including a polymer having arylamine repeating unit moiety represented by formula



wherein:

Ar, Ar_1, Ar_2, Ar_3, and Ar_4 are each individually aryl group of from 6 to 60 carbon atoms; or a heteroaryl group of from 4 to 60 carbons, or combinations thereof; or Ar_1 and Ar_2, Ar_3 and Ar_4, Ar_1 and Ar_4, Ar_2 and Ar_4 are connected through a chemical bond; and

X is a conjugated group having 2 to 60 carbon atoms.

The present invention provides light-emitting materials with a number of advantages that include good solubility, efficiency and stability. The emitting color of the polymer can be easily tuned by the incorporation of desired X group. Furthermore, other eletro-optical properties can also be tuned with X group. The low ionization potentials of the arylene diamine pendant side chain

enable the conjugated polymers of the present invention to be useful as hole injection materials as well.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 illustrates in cross-section a basic structure of an EL device;

FIG. 2 illustrates the EL spectra of EL devices fabricated from polymer 5, 28, and 58: ITO/PEDOT/polymer/CsF/Mg:Ag;

FIG. 3 illustrates the absorption (AB) and photoluminescence (PL) spectra of polymer 5 in solution and thin film; and

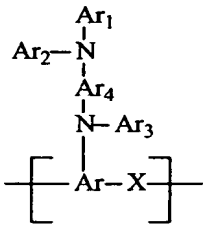
FIG. 4 illustrates voltage-current density-luminance characteristic of the EL device fabricated from polymer 5.

DETAILED DESCRIPTION OF THE INVENTION

The present invention provides polymers containing arylamine moieties with good solubility and efficiency, low driving voltage, and improved stability. Arylamine as a hole transport material in organic light-emitting devices was studied intensively due to its high hole transporting mobility, chemical and electronic stability. Arylamine moieties are strong electron donors that will improve the hole injection and transporting mobility of the polymer. Moreover, incorporating arylamine moieties into the polymer can enhance the solubility, improve polymer conductivity, and adjust polymer oxidation sensitivity. The low ionization potentials of the arylene diamine pendant side chain enable the conjugated polymers of the present invention to be useful as hole injection materials as well. Incorporation of group X described below into polymer has the following features:

1) to improve EL efficiency by achieving good balanced electron-hole injection and recombination of the charge carriers;
2) to further improve solubility of the polymer; and
3) to tune the emissive color of the polymer.

The present invention provides polymers containing arylamine moieties having the repeating unit represented by formula (I)



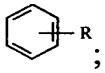
(I)

wherein:

Ar, Ar_1, Ar_2, Ar_3, and Ar_4 are each individually arylof from 6 to 60 carbon atoms; or a heteroarylof from 4 to 60 carbons, or combinations thereof; or Ar_1 and Ar_2, Ar_3 and Ar_4, Ar_1 and Ar_4, Ar_2 and Ar_4 are connected through a chemical bond.

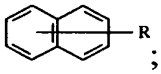
X is a conjugated group having 2 to 60 carbon atoms. The group can include vinylenes, ethynylenes, arylenes, heteroarylenes, arylene vinylenes, heteroarylene vinylenes and combinations thereof. X can include more than one conjugated group.

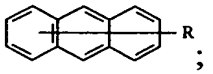
For example, Ar_1, Ar_2, Ar_3, and Ar_4 represent

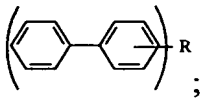


wherein:

R is a substituent being hydrogen, or alkyl, or alkenyl, or alkynyl, or alkoxy of from 1 to 40 carbon atoms; arylof from 6 to 60 carbon atoms; or heteroarylof from 4 to 60 carbons; or F, or Cl, or Br; or a cyano group; or a nitro group; or R is a group necessary to complete a fused aromatic or heteroaromatic ring;



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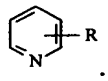
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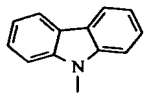
When Ar_1 and Ar_2, Ar_3 and Ar_4, Ar_1 and Ar_4, Ar_2 and Ar_4 are connected through a chemical bond, Ar_1 and Ar_2 together, Ar_3 and Ar_4 together, Ar_1 and Ar_4 together, Ar_2 and Ar_4 together contain 8 to 60 carbon atoms. For example, Ar_1 and Ar_2, Ar_3 and Ar_4, Ar_1 and Ar_4, Ar_2 and Ar_4 are connected by a chemical bond to form a group having $-\overset{Ar_1}{\underset{|}{N}}-Ar_2$, $-\overset{Ar_3}{\underset{|}{N}}-Ar_4$, $-\overset{Ar_1}{\underset{|}{N}}-Ar_4$, or $-\overset{Ar_1}{\underset{|}{N}}-Ar_4$ that includes the following carbazole and carbazole derivatives: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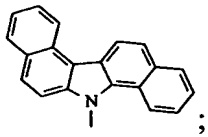
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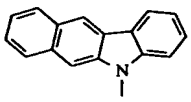
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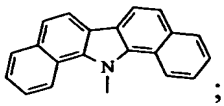
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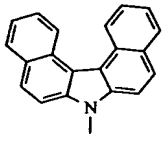
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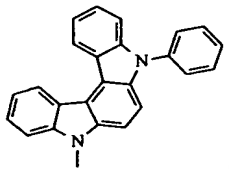
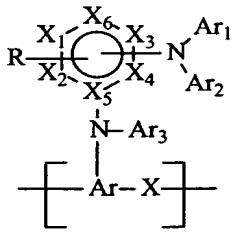


Preferably, Ar_4 is a six-member aryl or heteroaryl group and the conjugated polymers of the present invention is represented by the repeating unit of formula (II)

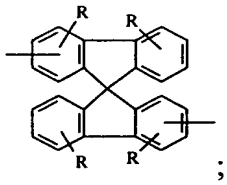


(II)

wherein:

X_1, X_2, X_3, X_4, X_5, and X_6 are individually the same or different and each include a moiety containing CH or N; and R is a substituent as defined above.

Ar represents the following groups:



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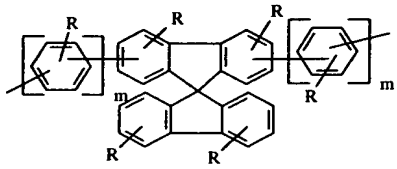
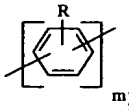


wherein: m is an integer from 1 to 4;



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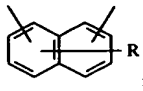
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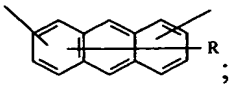
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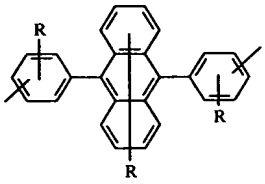
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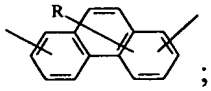
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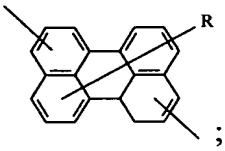
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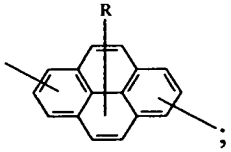
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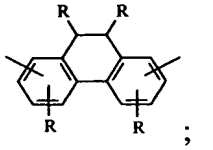
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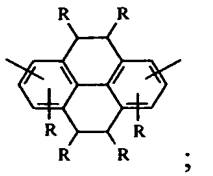
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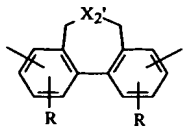
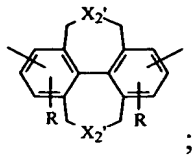


wherein: X_2' is S, Se, or O atom, SiR_2, or N-R; or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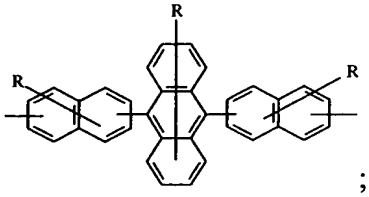
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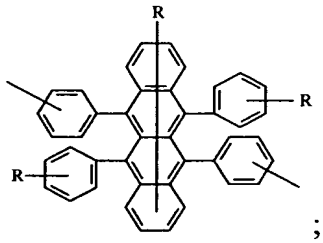
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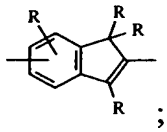
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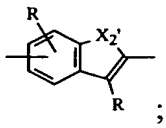
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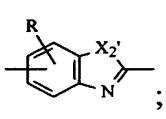
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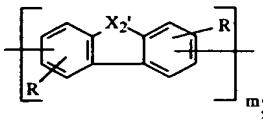
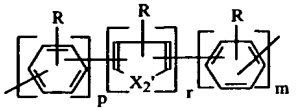
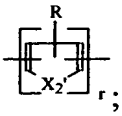
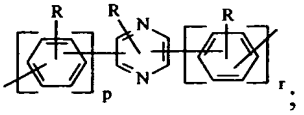
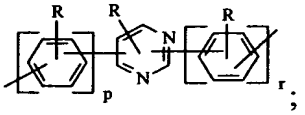


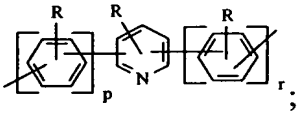


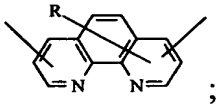
wherein p and r are integers from 1 to 4;

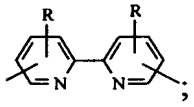


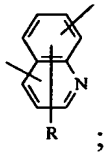


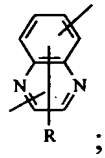








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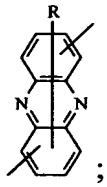
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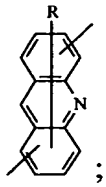
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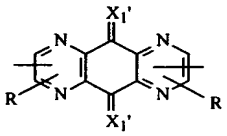
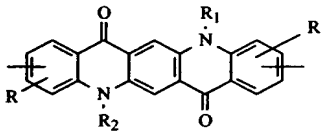
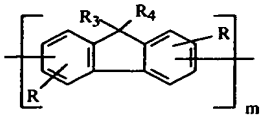


wherein: X_1' is an O atom or two cyano groups;

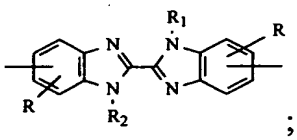


wherein: R_1 and R_2 are individually hydrogen, or alkyl of 1 to 40 carbon atoms, or arylcontaining 6 to 60 carbon atoms; or heteroarylcontaining 4 to 60 carbons; or F, Cl, or Br; or a cyano group; or a nitro group;



wherein: R_3 and R_4 are substituents each being individually hydrogen, or alkyl, or alkenyl, or alkynyl, or alkoxy of from 1 to 40 carbon atoms; arylof from

6 to 60 carbon atoms; or heteroarylof from 4 to 60 carbons; or F, Cl, or Br; or a cyano group; or a nitro group;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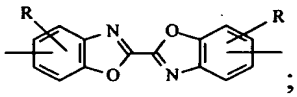
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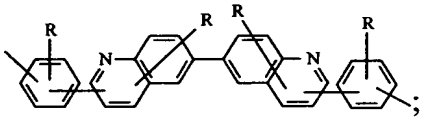
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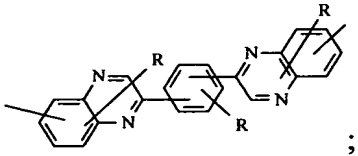
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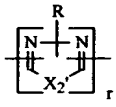
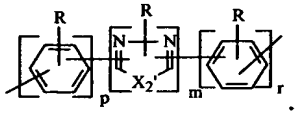

r; or


.

Ar can be one or the combination of more than one of the above groups.

X can be divided into the following groups.

Group I:

X is a vinylene, or ethynylene group of formula (II):

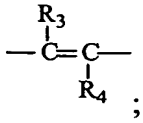
-W-

(II)

wherein:

W contains 2 to 40 carbon atoms, may also contains O, N, S, F, Cl, or Br, or Si atoms.

The following structures constitute specific examples of formula (II)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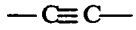


Group II:

X is a group containing two aryl or heteroaryl groups Ar_3 and Ar_4 connected by a linking group L_1 of formula (III):

$$—(Ar_7)—L_1—(Ar_8)—$$

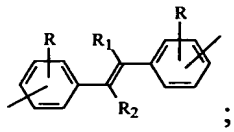
(III)

wherein:

Ar_7 and Ar_8 are substituted or unsubstituted aryl groups containing 6 to 60 carbon atoms, or heteroaryl groups containing 4 to 60 carbon atoms;

L_1 is a linking groups containing 0 to 40 carbon atoms, may contain N, Si, O, Cl, F, Br, or S atoms.

The following structures constitute specific examples of formula (III)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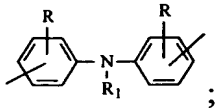
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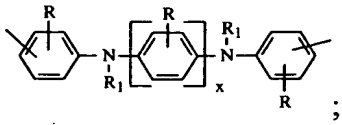
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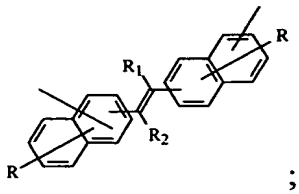
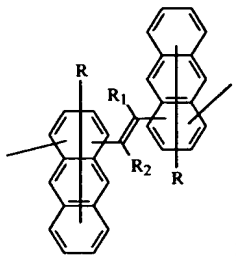
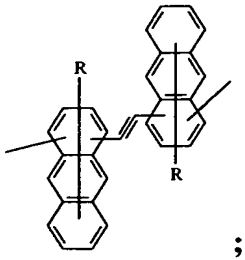
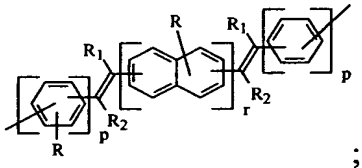
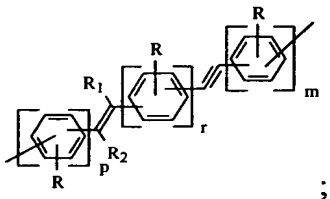
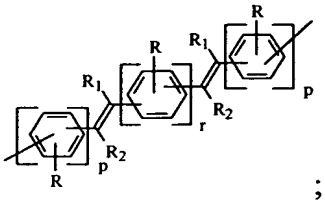

R
R1
R2
R
;

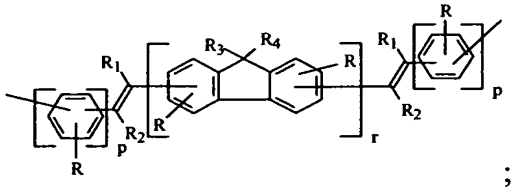

R
R1
R2
R
;


R
R
;


R
R1
R1
R2
R2
p
r
p
R
;


R
R
R1
R2
m
r
p
R
;


R
R
R1
R1
R2
p
r
R2
p
R
;



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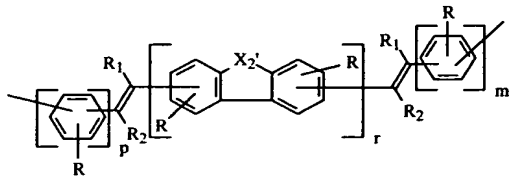
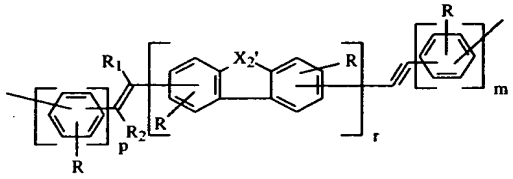


wherein: $X_{2'}$ is S, Se, or O atom, SiR_2, or N-R; or;



Group III:

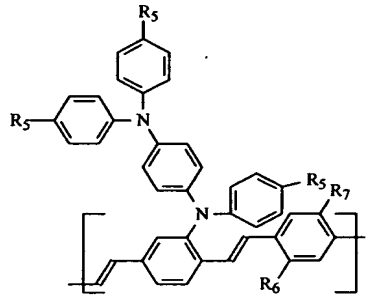
X is an aryl or heteroaryl group of formula (IV):

$$—Ar_9—$$

(IV)

wherein: Ar_9 is defined as Ar as noted above.

The following molecular structures constitute specific examples of preferred compounds satisfying the requirement of this invention:



polymer 1 $R_5 = R_6 =$ n-hexyl, $R_7 =$ 2-ethylhexyl

polymer 2 $R_5 = H$, $R_6 = R_7 =$ 3,7-dimethyloctyl

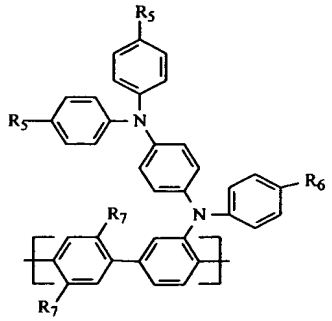
polymer 3 $R_5 =$ 4-(bis(4-methylphenyl)amino)phenyl, $R_6 = H$, $R_7 =$ t-butyl

polymer 4 R_5 = 4-(N-carbazole)phenyl, R_6 = n-decyl, R_7 = H

polymer 5 R_5 = H, R_6 = methoxy, R_7 = 3,7-dimethyloctyloxy

polymer 6 R_5 = R_6 = n-hexyloxy, R_7 = H

polymer 7 R_5 = R_6 = R_7 = n-hexyloxy



polymer 8 R_5 = R_6 = n-hexyl, R_7 = 2-ethylhexyl

polymer 9 R_5 = H, R_6 = R_7 = 3,7-dimethyloctyl

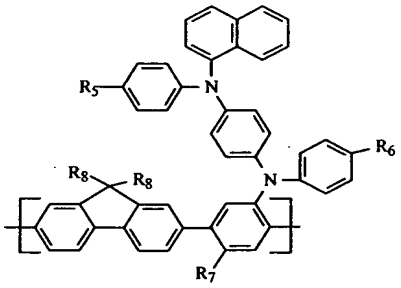
polymer 10 R_5 = 4-(bis(4-methylphenyl)amino)phenyl, R_6 = H, R_7 = t-butyl

polymer 11 R_5 = 4-(N-carbazole)phenyl, R_6 = n-decyl, R_7 = H

polymer 12 R_5 = n-hexyl, R_6 = R_7 = H

polymer 13 R_5 = R_6 = n-hexyloxy, R_7 = H

polymer 14 R_5 = R_6 = R_7 = n-hexyloxy



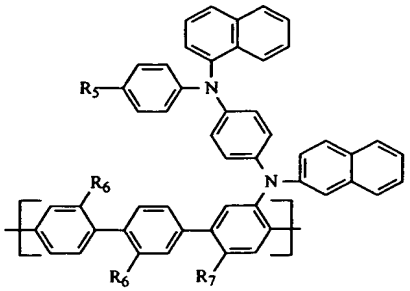
polymer 15 R_5 = R_6 = R_7 = R_8 = n-hexyl

polymer 16 R_5 = R_7 = H, R_6 = R_8 = 3,7-dimethyloctyl

polymer 17 R_5 = R_7 = H, R_6 = 4-(bis(4-methylphenyl)amino)phenyl, R_8 = n-hexyl

polymer 18 R_5 = 4-(N-carbazole)phenyl, R_6 = R_8 = n-decyl, R_7 = H

polymer 19 R_5 = n-hexyloxy, R_6 = R_7 = n-hexyl, R_8 = n-octyl



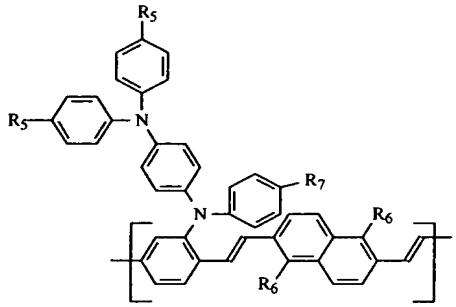
polymer 20 $R_5 = R_6 =$ n-hexyl, $R_7 =$ 2-ethylhexyl

polymer 21 $R_5 =$ methyl, $R_6 = R_7 =$ 3,7-dimethyloctyl

polymer 22 $R_5 =$ 4-(bis(4-methylphenyl)amino)phenyl, $R_6 =$ H, $R_7 =$ t-butyl

polymer 23 $R_5 =$ 4-(N-carbazole)phenyl, $R_6 =$ n-decyl, $R_7 =$ H

polymer 24 $R_5 =$ n-hexyl, $R_6 = R_7 =$ H

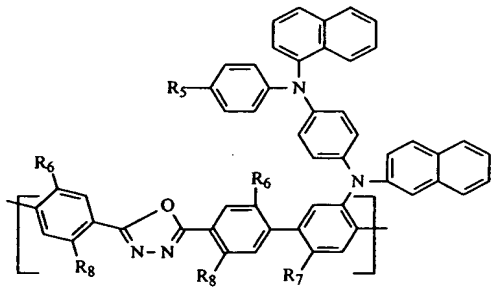


polymer 25 $R_5 = R_6 = R_7 =$ n-hexyl

polymer 26 $R_5 =$ n-hexyl, $R_6 =$ 3,7-dimethyloctyloxy, $R_7 =$ H

polymer 27 $R_5 = R_7 =$ methyl, $R_6 =$ 4-(bis(4-methylphenyl)amino)phenyl

polymer 28 $R_5 = R_7 =$ H, $R_6 =$ n-hexyloxy

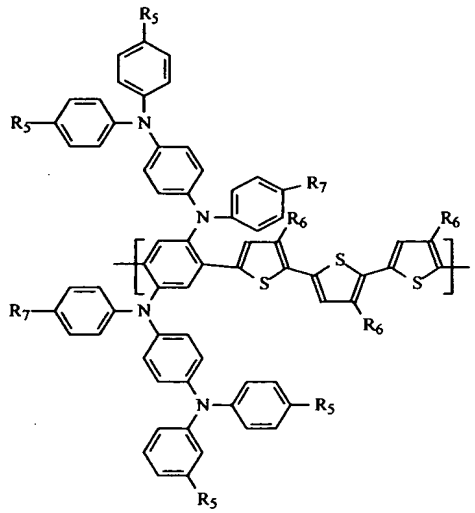


polymer 29 $R_5 = R_6 = R_7 = R_8 =$ n-hexyl

polymer 30 $R_5 =$ n-hexyl, $R_7 =$ H, $R_6 = R_8 =$ 3,7-dimethyloctyloxy

polymer 31 $R_5 = R_7 =$ H, $R_6 =$ 4-(bis(4-methylphenyl)amino)phenyl, $R_8 =$ H

polymer 32 R_5 = n-hexyloxy, R_6 = n-decyl, R_7 = R_8 = H



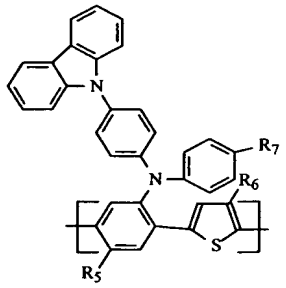
polymer 33 R_5 = R_6 = n-hexyl, R_7 = 2-ethylhexyl

polymer 34 R_5 = H, R_6 = R_7 = 3,7-dimethyloctyl

polymer 35 R_5 = R_7 = methyl, R_6 = 2-ethylhexyl

polymer 36 R_5 = R_6 = n-hexyl, R_7 = t-butyl

polymer 37 R_5 = R_7 = n-hexyloxy, R_6 = 2-ethylhexyl



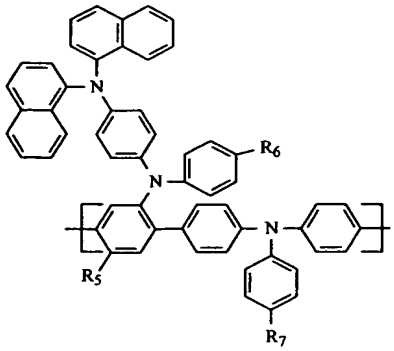
polymer 38 R_5 = R_6 = n-hexyl, R_7 = 2-ethylhexyl

polymer 39 R_5 = H, R_6 = n-hexyl, R_7 = t-butyl

polymer 40 R_5 = R_7 = methyl, R_6 = 4-t-butylphenyl

polymer 41 R_5 = R_6 = n-hexyl, R_7 = H

polymer 42 R_5 = R_7 =H, R_6 = 2-ethylhexyl



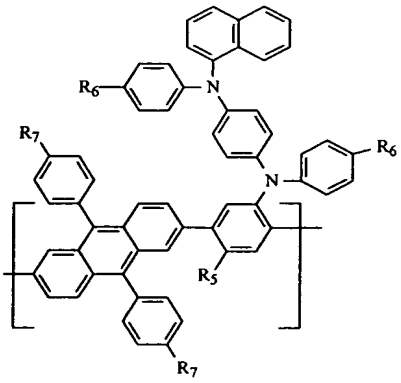
polymer 43 R_5 = n-hexyl, R_6 = H, R_7 = n-decyl

polymer 44 $R_5 = R_7$ = 2-ethylhexyl, R_6 = H

polymer 45 $R_5 = R_6$ = n-hexyl, R_7 = (4-carbazole)phenyl

polymer 46 R_5 = n-hexyloxy, R_6 = H, R_7 = 3,7-dimethyloctyl

polymer 47 $R_5 = R_6$ = n-hexyl, R_7 = trifluoromethyl



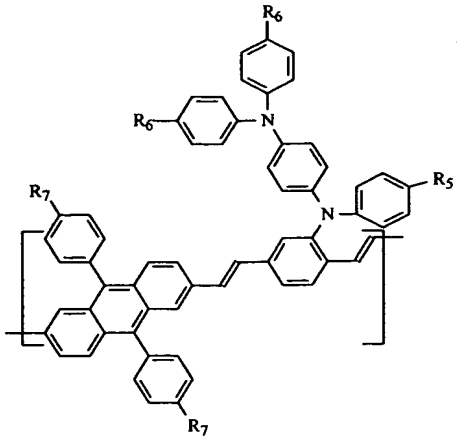
polymer 48 R_5 = n-hexyl, R_6 = H, R_7 = t-butyl

polymer 49 $R_5 = R_7$ = 3,7-dimethyloctyl, R_6 = H

polymer 50 $R_5 = R_6 = R_7$ = n-hexyl

polymer 51 R_5 = H, R_7 = n-hexyloxy, R_6 = diphenylamino

polymer 52 $R_5 = R_6$ = H, R_7 = trifluoromethyl



polymer 53 R_5 = n-hexyl, R_6 = H, R_7 = t-butyl

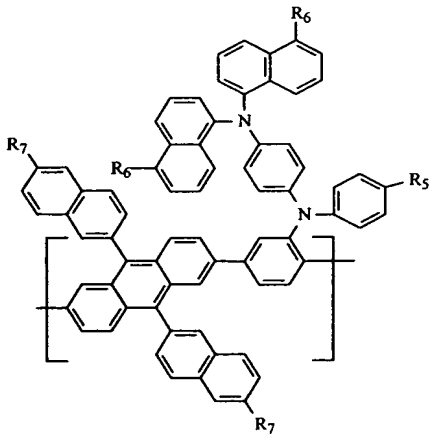
polymer 54 R_5 = n-hexyl, R_6 = R_7 = 2-ethylhexyl

polymer 55 R_5 = methyl, R_6 = H, R_7 = 3,7-dimethyloctyl

polymer 56 R_5 = R_6 = n-hexyl, R_7 = (4-carbazole)phenyl

polymer 57 R_5 = n-hexyloxy, R_6 = H, R_7 = diphenylamino

polymer 58 R_5 = R_6 = H, R_7 = 2-ethylhexyloxy



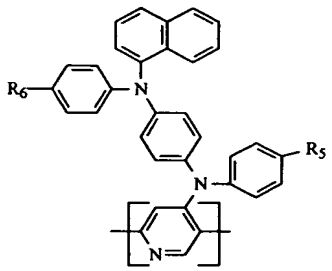
polymer 59 R_5 = R_6 = R_7 = n-hexyl

polymer 60 R_5 = n-hexyl, R_6 = H, R_7 = 2-ethylhexyloxy

polymer 61 R_5 = 4-(bis(4-methylphenyl)amino)phenyl, R_6 = n-hexyl, R_7 = n-decyl

polymer 62 R_5 = H, R_6 = methyl, R_7 = 3,7-dimethyloctyl

polymer 63 R_5 = R_7 = n-hexyloxy, R_6 = H



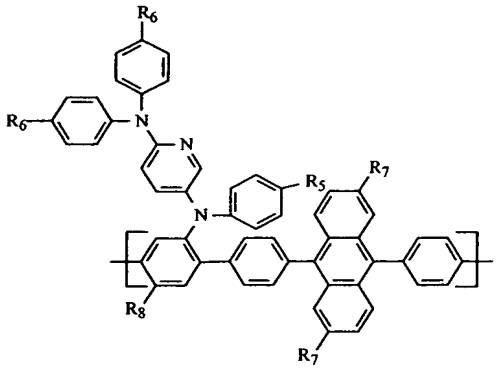
polymer 64 $R_5 = R_6 =$ n-hexyl

polymer 65 $R_5 =$ n-hexyl, $R_6 = H$

polymer 66 $R_5 =$ 4-(bis(4-methylphenyl)amino)phenyl, $R_6 =$ 2-ethylhexyl

polymer 67 $R_5 = R_6 =$ n-hexyloxy

polymer 68 $R_5 = H$, $R_6 =$ n-hexyloxy



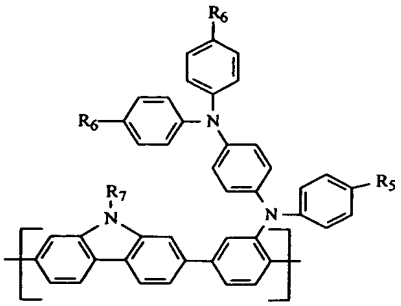
polymer 69 $R_5 = R_6 =$ n-hexyl, $R_7 = R_8 =$ 2-ethylhexyloxy

polymer 70 $R_5 =$ n-hexyl, $R_6 = R_7 = H$, $R_8 =$ t-butyl

polymer 71 $R_5 = R_6 = H$, $R_7 = R_8 =$ 4-(bis(4-methylphenyl)amino)phenyl

polymer 72 $R_5 =$ n-hexyloxy, $R_6 = R_8 = H$, $R_7 =$ 2-ethylhexyl

polymer 73 $R_5 = H$, $R_6 =$ phenyl, $R_7 = R_8 =$ 3,7-dimethyloctyl



polymer 74 $R_5 =$ n-hexyl, $R_6 = H$, $R_7 =$ (4-t-butyl)phenyl

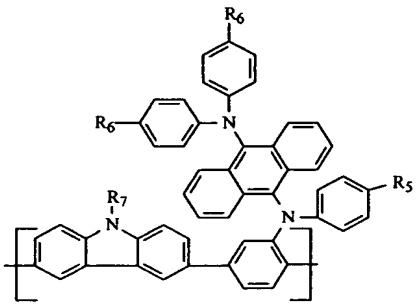
polymer 75 R_5 = n-hexyl, R_6 = H, R_7 = 2-ethylhexyl

polymer 76 $R_5 = R_7$ = 4-(bis(4-methylphenyl)amino)phenyl, R_6 = n-hexyl

polymer 77 $R_5 = R_6$ = H, R_7 = 2-ethylhexyl

polymer 78 R_5 = n-hexyloxy, R_6 = H, R_7 = t-butyl

polymer 79 $R_5 = R_6$ = trifluoromethyl, R_7 = n-hexyl

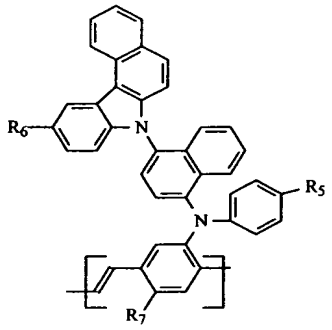


polymer 80 R_5 = n-hexyl, R_6 = H, R_7 = (4-t-butyl)phenyl

polymer 81 $R_5 = R_6$ = H, R_7 = 3,7-dimethyloctyl

polymer 82 $R_5 = R_6$ = n-hexyl, R_7 = 2-ethylhexyl

polymer 83 R_5 = n-hexyloxy, R_6 = H, R_7 = t-butyl



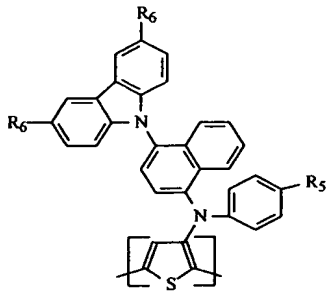
polymer 84 $R_5 = R_6 = R_7$ = n-hexyl

polymer 85 $R_5 = R_6$ = n-hexyloxy, R_7 = H

polymer 86 $R_5 = R_7$ = H, R_6 = n-octyl

polymer 87 R_5 = n-decyl, R_6 = phenyl, R_7 = H

polymer 88 R_5 = n-hexyloxy, $R_6 = R_7$ = n-hexyl

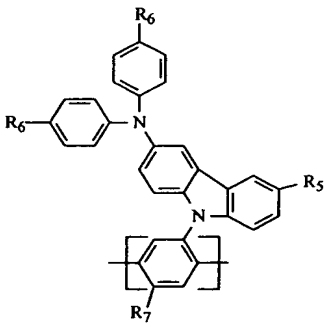


polymer 89 R_5 = n-hexyl, R_6 = H

polymer 90 $R_5 = R_6$ = n-hexyl

polymer 91 R_5 = H, R_6 = 2-ethylhexyloxy

polymer 92 R_5 = H, R_6 = 2-ethylhexyl

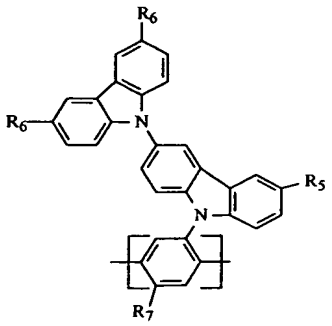


polymer 93 $R_5 = R_7$ = n-hexyl, R_6 = H

polymer 94 $R_5 = R_6$ = methyl, R_7 = n-decyl

polymer 95 $R_5 = R_6 = R_7$ = n-hexyl

polymer 96 $R_5 = R_6 = R_7$ = n-hexyloxy

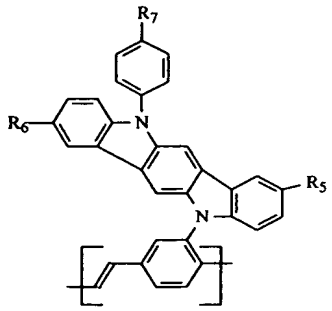


polymer 97 R_5 = n-hexyl, R_6 = H, R_7 = (4-t-butyl)phenyl

polymer 98 R_5 = H, R_6 = n-hexyl, R_7 = 2-ethylhexyl

polymer 99 $R_5 = R_6$ = H, R_7 = 4-decylphenyl)

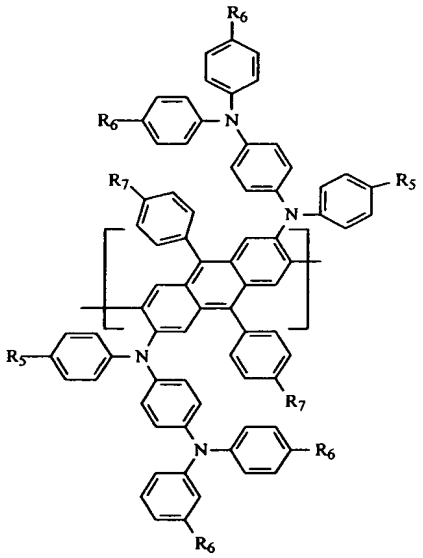
polymer 100 $R_5 = R_6$ = n-hexyl, R_7 = 2-ethylhexyloxy



polymer 101 R_5 = n-hexyl, R_6 = H, R_7 = (4-t-butyl)phenyl

polymer 102 $R_5 = R_6$ = n-hexyl, R_7 = 2-ethylhexyl

polymer 103 $R_5 = R_7$ = n-hexyloxy, R_6 = H

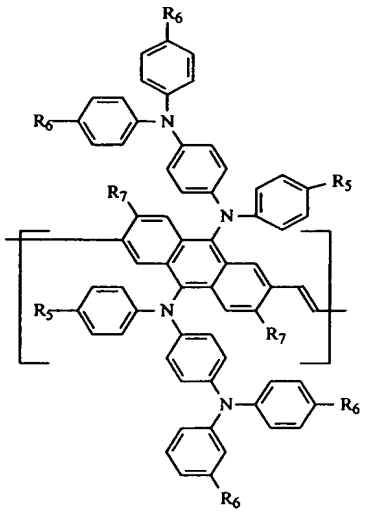


polymer 104 $R_5 = R_6 = R_7$ = n-hexyl

polymer 105 $R_5 = R_6$ = H, R_7 = 4-octylphenyl

polymer 106 $R_5 = R_6$ = methyl, R_7 = 2-ethylhexyloxy

polymer 107 $R_5 = R_6 = R_7$ = n-hexyloxy

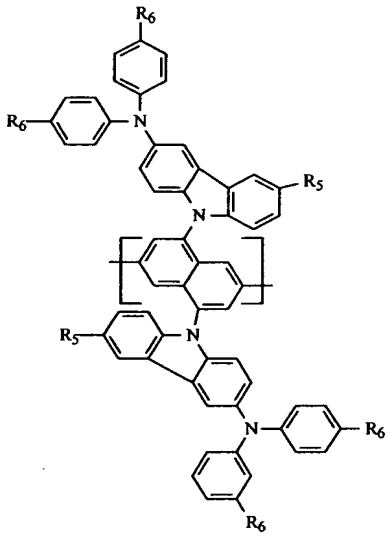


polymer 108 $R_5 = R_6 =$ 3,7-dimethyloctyl, $R_7 = H$

polymer 109 $R_5 = H$, $R_6 =$ 4-t-butylphenyl, $R_6 =$ 2-ethylhexyl

polymer 110 $R_5 = R_6 =$ n-hexyloxy, $R_7 = H$

polymer 111 $R_5 =$ n-hexyloxy, $R_6 =$ 2-ethylhexyl, $R_7 =$ t-butyl

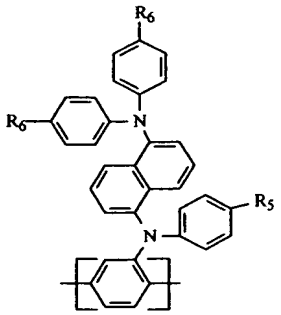


polymer 112 $R_5 = R_6 =$ n-hexyl

polymer 113 $R_5 = R_6 =$ n-hexyloxy

polymer 114 $R_5 = H$, $R_6 =$ n-octyl

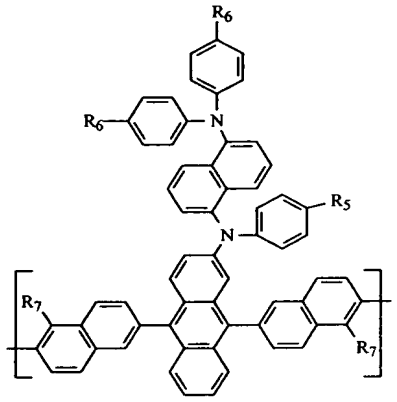
polymer 115 $R_5 =$ methyl, $R_6 =$ 4-hexylphenyl



polymer 116 R_5 = H, R_6 = n-hexyloxy

polymer 117 R_5 = n-hexyl R_6 = 4-(t-butylphenyl)

polymer 118 R_5 = R_6 = 2-ethylhexyl

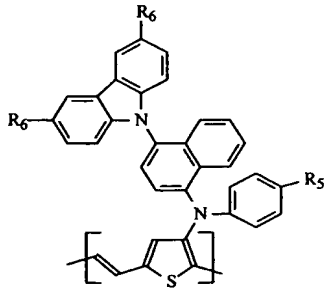


polymer 119 R_5 = n-hexyl, R_6 = H, R_7 = 2-ethylhexyl

polymer 120 R_5 = methyl, R_6 = n-hexyloxy, R_7 = 4-(t-butylphenyl)

polymer 121 R_5 = R_6 = n-hexyl, R_7 = H

polymer 122 R_5 = H, R_6 = n-hexyl, R_7 = 3,7-dimethyloctyl

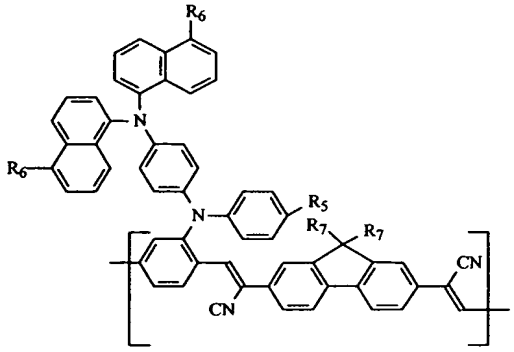


polymer 123 R_5 = H, R_6 = n-hexyl

polymer 124 R_5 = R_6 = n-hexyloxy

polymer 125 R_5 = 4-(bisphenylamino)phenyl, R_6 = 2-ethylhexyl

polymer 126 R_5 = n-decyloxy, R_6 = H

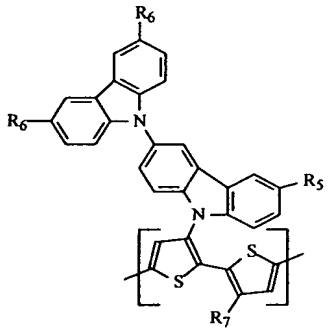


polymer 127 $R_5 = R_6 = R_7$ = n-hexyl

polymer 128 R_5 = methyl, R_6 = n-hexyloxy, R_7= n-hexyl

polymer 129 R_5 = H, R_6 = n-octyl, R_7 = 4-(bis(4-methylphenyl)amino)phenyl

polymer 130 $R_5 = R_6$ = H, R_7 = 3,7-dimethyloctyl

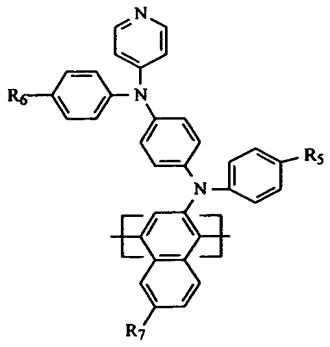


polymer 131 $R_5 = R_7$ = H, R_6 = n-hexyl

polymer 132 $R_5 = R_7$ = 4-(bis(4-methylphenyl)amino)phenyl, R_6 = n-decyl

polymer 133 $R_5 = R_6$ = n-hexyl, R_7 = H

polymer 134 R_5 = H, R_6 = n-hexyloxy, R_7 = n-hexyl

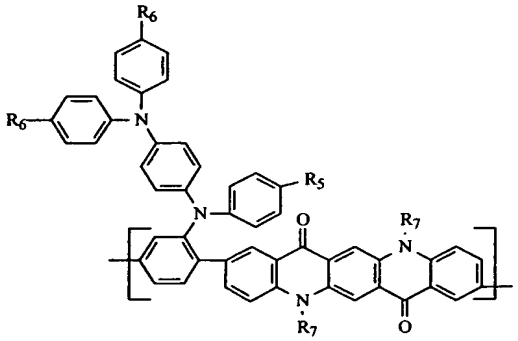


polymer 135 $R_5 = R_6 = R_7$ = n-hexyl

polymer 136 $R_5 = R_7$ = 4-(bis(4-hexylphenyl)amino)phenyl, R_6 = H

polymer 137 $R_5 = R_6$ = n-hexyl, R_7 = 2-ethylhexyloxy

polymer 138 $R_5 = R_7$ = H, R_6 = n-hexyloxy



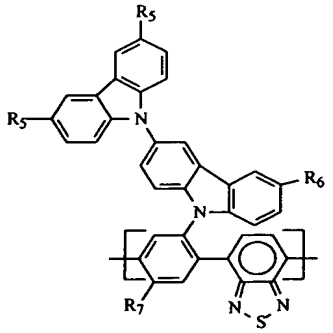
polymer 139 R_5 = H, R_6 = n-hexyl, R_7 = (4-t-butyl)phenyl

polymer 140 $R_5 = R_6$ = 2-ethylhexyl, R_7 = 4-(bis(4-methylphenyl)amino)phenyl

polymer 141 $R_5 = R_6 = R_7$ = 2-ethylhexyl

polymer 142 $R_5 = R_6$ = n-hexyloxy, R_7 = t-butyl

polymer 143 $R_5 = R_6$ = 4-hexylphenyl, R_7 = trifluoromethyl



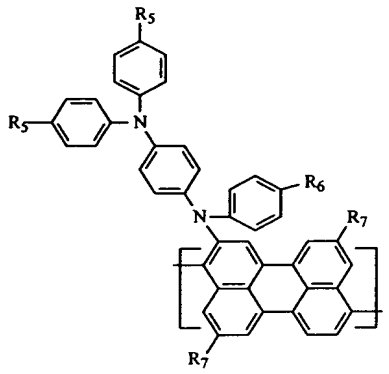
polymer 144 R_5 = n-hexyl, R_6 = H, R_7 = (4-t-butyl)phenyl

polymer 145 $R_5 = R_6 = R_7$ = n-hexyloxy

polymer 146 $R_5 = R_6$ = n-hexyl, R_7 = 2-ethylhexyl

polymer 147 R_5 = n-hexyloxy, $R_6 = R_7$ = 2-ethylhexyl

polymer 148 $R_5 = R_6 = R_7$ = n-hexyl



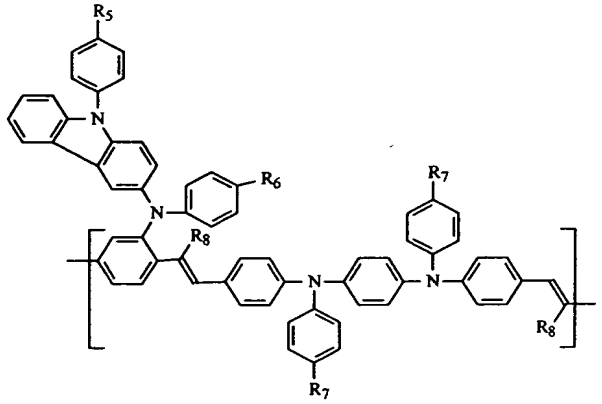
polymer 149 R_5 = n-hexyl, R_6 = H, R_7 = (4-decyl)phenyl

polymer 150 $R_5 = R_6 = R_7$ = n-hexyl

polymer 151 $R_5 = R_6$ = n-hexyl, R_7 = 2-ethylhexyloxy

polymer 152 R_5 = n-hexyloxy, R_6 = 2-ethylhexyl, R_7 = H

polymer 153 $R_5 = R_6$ = n-octyl, R_7 = trifluoromethyl

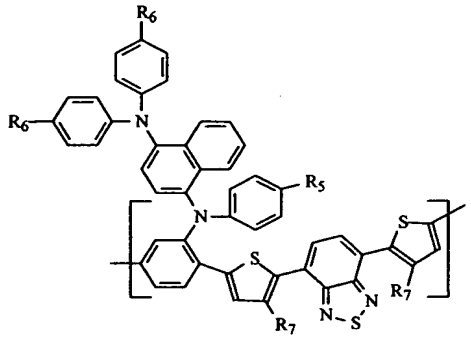


polymer 154 $R_5 = R_6$ = n-hexyl, R_7 = t-butyl, R_8 = H

polymer 155 R_5 = 2-ethylhexyl, R_6 = n-hexyl, R_7 = 4-t-butylphenyl, R_8 = CN

polymer 156 $R_5 = R_6$ = n-hexyloxy, R_7 = t-butyl, R_8 = phenyl

polymer 157 R_5 = n-hexyl, R_6 = H, R_7 = (4-diphenylamino)phenyl, R_8 = CN

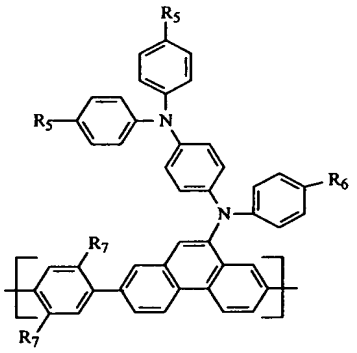


polymer 158 $R_5 = R_6 = R_7 =$ n-hexyl

polymer 159 $R_5 = R_6 =$ 2-ethylhexyloxy, $R_7 = H$

polymer 160 $R_5 =$ n-hexyoxy, $R_6 =$ n-hexyl, $R_7 = H$

polymer 161 $R_5 =$ n-hexyl, $R_6 = H$, $R_7 =$ (4-diphenylamino)phenyl

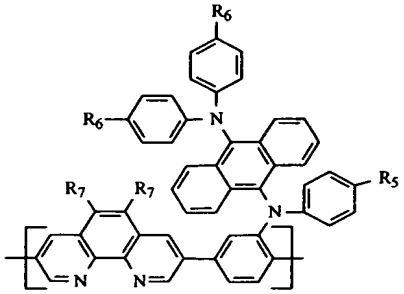


polymer 162 $R_5 = R_6 = R_7 =$ n-hexyl

polymer 163 $R_5 =$ 2-ethylhexyl, $R_6 =$ n-hexyloxy, $R_7 = H$

polymer 164 $R_5 = R_6 = R_7 =$ n-hexyloxy

polymer 165 $R_5 =$ n-hexyl, $R_6 = H$, $R_7 =$ (4-diphenylamino)phenyl

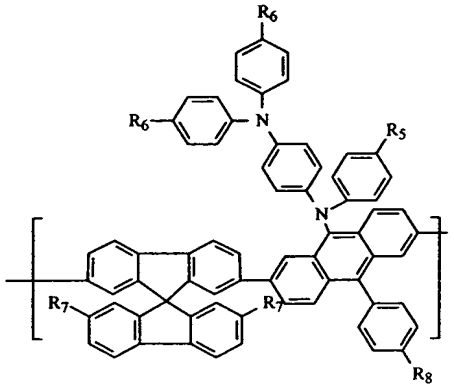


polymer 166 $R_5 = R_6 =$ n-hexyl, $R_7 =$ phenyl

polymer 167 R_5 = 2-ethylhexyl, R_6 = n-hexyloxy, R_7 = H

polymer 168 $R_5 = R_6 = R_7$ = 3,7-dimethyloctyloxy

polymer 169 R_5 = methyl, R_6 = 3,7-dimethyloctyl, R_7 = (4-diphenylamino)phenyl

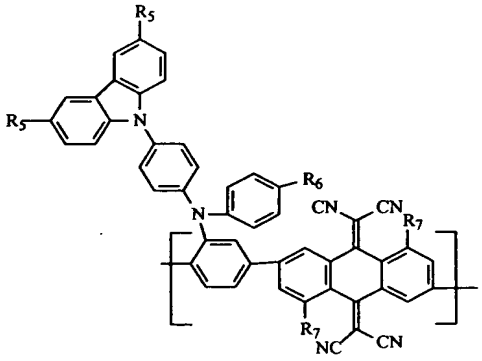


polymer 170 $R_5 = R_6$ = n-hexyl, R_7 = H, R_8 = 2-ethylhexyloxy

polymer 171 $R_5 = R_6$ = n-hexyloxy, R_7 = 2-ethylhexyloxy, R_8 = H

polymer 172 $R_5 = R_7$ = H, R_6 = 2-ethylhexyl, R_8 = 4-(bis(4-methylphenyl)amino)phenyl

polymer 173 R_5 = n-hexyl, $R_6 = R_8$ = 2-ethylhexyl

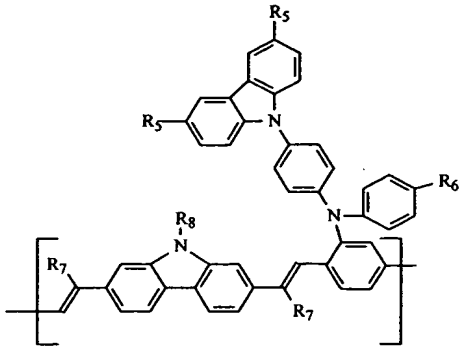


polymer 174 $R_5 = R_7$ = n-hexyloxy, R_6 = H

polymer 175 $R_5 = R_6$ = n-hexyl, R_7 = 3,7-dimethyloctyloxy

polymer 176 R_5 = n-octyl, R_6 = methyl, R_7 = 4-hexylphenyl

polymer 177 R_5 = trifluoromethyl, R_6 = t-butylphenyl, R_7 = 2-ethylhexyl

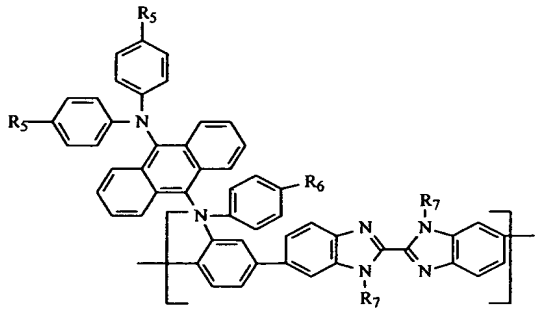


polymer 178 $R_5 = R_6$ = n-hexyl, R_7 = phenyl, R_8 = 2-ethylhexyl

polymer 179 R_5 = n-hexyl, R_6 = H, R_7 = CN, R_8 = 3,7-dimethyloctyloxy

polymer 180 R_5 = n-hexyloxy, $R_6 = R_8$ = 3,7-dimethyloctyl, R_7 = H

polymer 181 R_5 = 2-ethylhexyl, R_6 = n-hexyl, R_7 = H, R_8 = 4-t-butylphenyl

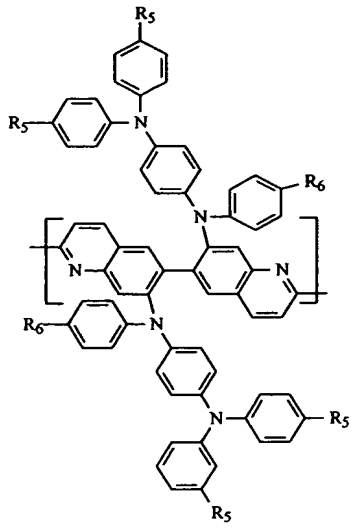


polymer 182 $R_5 = R_6 = R_7$ = n-hexyl

polymer 183 R_5 = n-decyl, R_6 = H, R_7 = (4-diphenylamino)phenyl

polymer 184 $R_5 = R_6$ = n-hexyloxy, R_7 = 4-t-butylphenyl

polymer 185 R_5 = 4-t-butylphenyl, R_6 = methyl, R_7 = 2-ethylhexyl

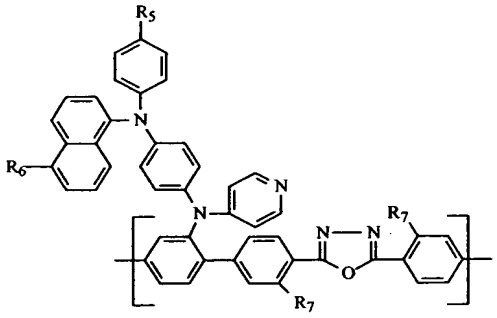


polymer 186 $R_5 = R_6$ = n-hexyl

polymer 187 $R_5 = R_6$ = n-hexyloxy

polymer 188 R_5 = 2-ethylhexyl R_6 = H

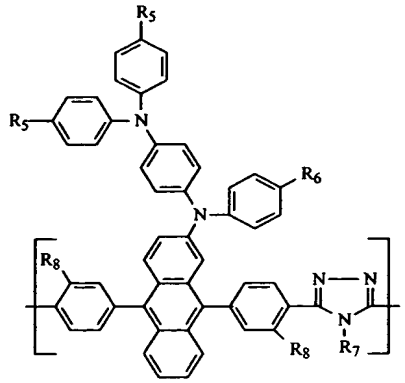
polymer 189 R_5 = 4-hexyloxyphenyl, R_6 = methyl



polymer 190 R_5 = n-hexyl, R_6 = H, R_7 = (4-diphenylamino)phenyl

polymer 191 $R_5 = R_7$ = n-hexyloxy, R_6 = H

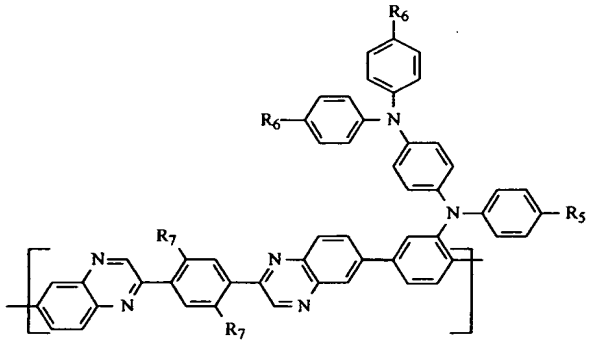
polymer 192 $R_5 = R_6 = R_7$ = n-hexyl



polymer 193 $R_5 = R_7$ = 2-ethylhexyl, R_6 = H, R_8 = 2-ethylhexyloxy

polymer 194 $R_5 = R_6$ = n-hexyloxy, R_7 = 3,7-dimethyloctyloxy, R_8 = H

polymer 195 R_5 = methyl, R_6 = (4-diphenylamino)pheny,l $R_7 = R_8$ = 3,7-dimethyloctyl

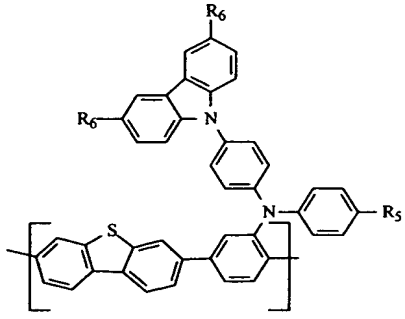


polymer 196 $R_5 = R_6$ = n-hexyl, R_7 = t-butyl

polymer 197 $R_5 = R_6$ = 2-ethylhexyl, R_7 = H

polymer 198 $R_5 = R_7$ = n-octyloxy, R_6 = methyl,

polymer 199 R_5 = n-hexyl, R_6 = H, R_7 = (4-diphenylamino)phenyl

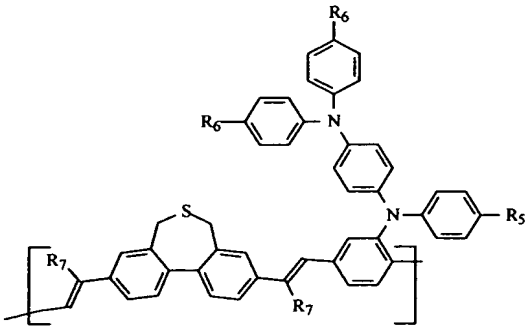


polymer 200 $R_5 = R_6$ = n-hexyl

polymer 201 $R_5 = R_6 =$ 2-ethylhexyloxy

polymer 202 $R_5 =$ n-hexyloxy, $R_6 =$ (4-diphenylamino)phenyl

polymer 203 $R_5 = R_6 =$ 4-hexylphenyl

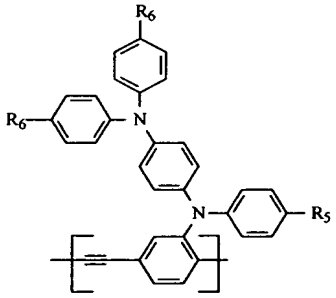


polymer 204 $R_5 = R_6 =$ n-hexyl, $R_7 = H$

polymer 205 $R_5 =$ methyl, $R_6 =$ 2-ethylhexyloxy, $R_7 =$ phenyl

polymer 206 $R_5 = R_6 =$ n-hexyloxy, $R_7 = CN$

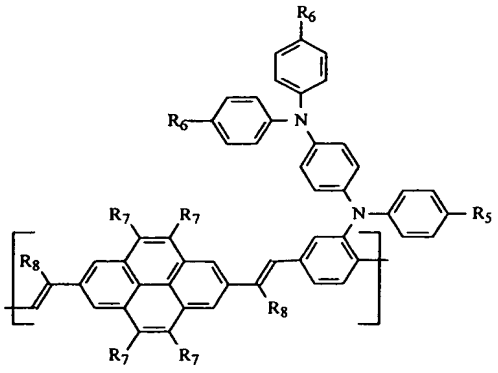
polymer 207 $R_5 =$ (4-diphenylamino)phenyl, $R_6 =$ 4-hexylphenyl, $R_7 = CN$



polymer 208 $R_5 = R_6 =$ n-hexyl

polymer 209 $R_5 =$ 4-(bis(4-methylphenyl)amino)phenyl, $R_6 =$ hexyloxy

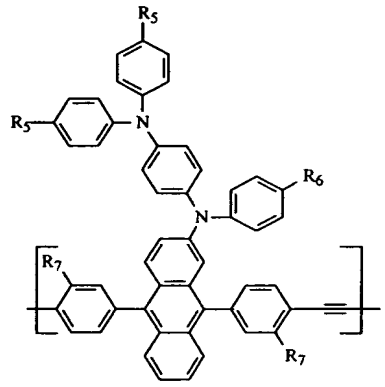
polymer 210 $R_5 = R_6 =$ n-hexyloxy



polymer 211 $R_5 = R_6$ = n-hexyloxy, R_7 = 2-ethylhexyl, R_8 = phenyl

polymer 212 $R_5 = R_6$ = n-hexyl, R_7 = 2-ethylhexyloxy, R_8 = H

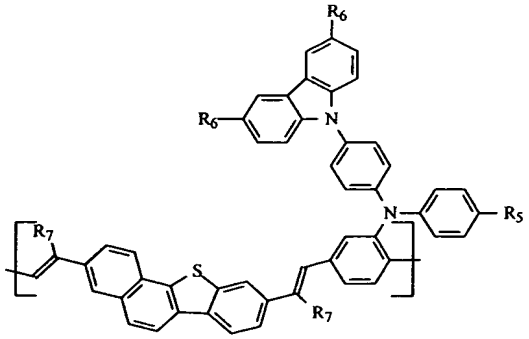
polymer 213 R_5 = 4-hexylphenyl, R_6 = methyl, R_7 = n-octyl, R_8 = CN



polymer 214 $R_5 = R_6 = R_7$ = n-hexyloxy

polymer 215 R_5 = n-hexyloxy, R_6 = 2-ethylhexyl, R_7 = H

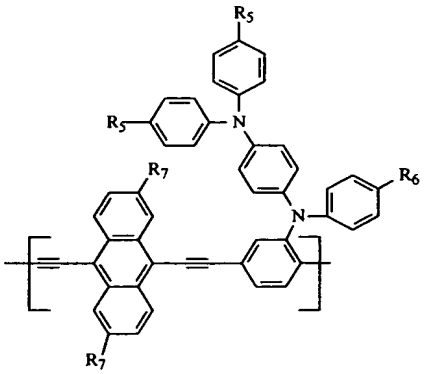
polymer 216 $R_5 = R_6 = R_7$ = n-hexyl



polymer 217 $R_5 = R_6$ = 2-ethylhexyloxy, R_7 = H

polymer 218 R_5 = methyl, R_6 = n-hexyl, R_7 = CN

polymer 219 R_5 = (4-diphenylamino)phenyl, R_6 = 2-ethylhexyl, R_7 = phenyl



polymer 220 $R_5 = R_6 = R_7$ = n-hexyl

polymer 221 $R_5 = R_6 = R_7$ = hexyloxy

polymer 222 $R_5 = R_7$ = 2-ethylhexyl, R_6 = di-tolylamino

The specific molecular structures can be the combination of any of the above drawn structures.

The conjugated polymers comprising arylamine structure (I) can be synthesized using known methods. The polymerization method and the molecular weights of the resulting polymers used in the present invention are not necessary to be particularly restricted. The molecular weights of the polymers are at least 1000, and preferably at least 2000. The polymers may be prepared by condensation polymerizations, such as coupling reactions including Pd-catalyzed Suzuki coupling, Stille coupling or Heck coupling, or Ni-mediated Yamamoto coupling, or by other condensation methods such as Wittig reaction, or Horner-Emmons reaction, or Knoevenagel reaction, or dehalogenation of dibenzyl halides. According to the present invention, the above mentioned polymers were prepared by a Horner-Emmons reaction between an aromatic dicarboxyaldehyde and a diphosphate, or a Knoevenagel reaction using an aromatic dicarboxyaldehyde and a dicyano compound in the presence of a strong base such as potassium t-butoxide or sodium hydride.

Suzuki coupling reaction was first reported by Suzuki et al on the coupling of aromatic boronic acid derivatives with aromatic halides (Suzuki, A. et al *Synthetic Comm.* 1981, *11(7)*, 513). Since then, this reaction has been widely used to prepared polymers for various applications (Ranger, M. et al *Macromolecules* 1997, *30*, 7686). The reaction involves the use of a palladium-

based catalyst such as a soluble Pd compound either in the state of Pd (II) or Pd (0), a base such as an aqueous inorganic alkaline carbonate or bicarbonate, and a solvent for the reactants and/or product. The preferred Pd catalyst is a Pd (0) complex such as $Pd(PPh_3)_4$ or a Pd (II) salt such as $Pd(PPh_3)_2Cl_2$ or $Pd(OAc)_2$ with a tertiary phosphine ligand, and used in the range of 0.01-10 mol% based on the functional groups of the reactants. Polar solvents such as THF and non-polar solvents toluene can be used however, the non-polar solvent is believed to slow down the reaction. Modified processes were reported to prepare conjugated polymers for EL devices from the Suzuki coupling of aromatic halides and aromatic boron derivatives (Inbasekaran, M. et al U.S. Patent No. 5,777,070 (1998); Towns, C. R. et al. PCT WO00/53656, 2000). A variation of the Suzuki coupling reaction replaces the aromatic halide with an aromatic trifluoromethanesulfonate (triflate) (Ritter, K. Synthesis, 1993, 735). Aromatic triflates are readily prepared from the corresponding phenol derivatives. The advantages of using aromatic triflates are that the phenol derivatives are easily accessible and can be protected/deprotected during complex synthesis. For example, aromatic halides normally would react under various coupling conditions to generate unwanted by-product and lead to much more complicated synthetic schemes. However, phenol derivatives can be easily protected by various protecting groups that would not interfere with functional group transformation and be deprotected to generate back the phenol group which then can be converted to triflates. The diboron derivatives can be prepared from the corresponding dihalide or ditriflate.

The synthetic schemes of the polymers according to the present invention are illustrated in Schemes 1-3.

The process of the invention provides conjugated polymers particularly useful for an optical device. The optical device may comprise a luminescent device such as an EL device in which the polymers of the present invention is deposited between a cathode and an anode. The polymers can be deposited as thin film by vapor deposition or thermal transfer method or from a solution by spin-coating, spray-coating, dip-coating, roller-coating, or ink jet delivery. The thin film may be supported by substrate directly, preferably a

transparent substrate, or supported by the substrate indirectly where there is one or more inter layers between the substrate and thin film. The thin film can be used as emitting layer or charge carrier transporting layer.

General EL device architecture:

The present invention can be employed in most organic EL device configurations. These include very simple structures including a single anode and cathode to more complex devices, such as passive matrix displays comprised of orthogonal arrays of anodes and cathodes to form pixels, and active-matrix displays where each pixel is controlled independently, for example, with thin film transistors (TFTs).

There are numerous configurations of the organic layers wherein the present invention can be successfully practiced. A typical structure is shown in FIG. 1 and includes a substrate 101, an anode 103, a hole-injecting layer 105, a hole-transporting layer 107, a light-emitting layer 109, an electron-transporting layer 111, and a cathode 113. These layers are described in detail below. This figure is for illustration only and the individual layer thickness is not scaled according to the actual thickness. Note that the substrate 101 may alternatively be located adjacent to the cathode 113, or the substrate may actually constitute the anode 103 or cathode 113. The organic layers between the anode 103 and cathode 113 are conveniently referred to as the organic EL element.

The anode 103 and cathode 113 of the OLED are connected to a voltage/current source 250 through electrical conductors 260. The OLED is operated by applying a potential between the anode 103 and cathode 113 such that the anode 103 is at a more positive potential than the cathode 113. Holes are injected into the organic EL element from the anode 103 and electrons are injected into the organic EL element at the anode 103. Enhanced device stability can sometimes be achieved when the OLED is operated in an AC mode where, for some time period in the cycle, the potential bias is reversed and no current flows. An example of an AC driven OLED is described in U.S. Patent No. 5,552,678.

Substrate:

The OLED device of this invention is typically provided over a supporting substrate 101 where either the cathode 113 or anode 103 can be in

contact with the substrate 101. The electrode in contact with the substrate 101 is conveniently referred to as the bottom electrode. Conventionally, the bottom electrode is the anode 103, but this invention is not limited to that configuration. The substrate 101 can either be light transmissive or opaque, depending on the intended direction of light emission. The light transmissive property is desirable for viewing the EL emission through the substrate 101. Transparent glass or plastic is commonly employed in such cases. The substrate 101 may be a complex structure comprising multiple layers of materials. This is typically the case for active matrix substrates wherein TFTs are provided below the EL layers. It is still necessary that the substrate, at least in the emissive pixilated areas, be comprised of largely transparent materials such as glass or polymers. For applications where the EL emission is viewed through the top electrode, the transmissive characteristic of the bottom support is immaterial, and therefore can be light transmissive, light absorbing or light reflective. Substrates for use in this case include, but are not limited to, glass, plastic, semiconductor materials, silicon, ceramics, and circuit board materials. Again, the substrate may be a complex structure comprising multiple layers of materials such as found in active matrix TFT designs. Of course it is necessary to provide in these device configurations a light-transparent top electrode.

Anode:

When EL emission is viewed through anode 103, the anode 103 should be transparent or substantially transparent to the emission of interest. Common transparent anode materials used in this invention are indium-tin oxide (ITO), indium-zinc oxide (IZO) and tin oxide, but other metal oxides can work including, but not limited to, aluminum- or indium-doped zinc oxide, magnesium-indium oxide, and nickel-tungsten oxide. In addition to these oxides, metal nitrides, such as gallium nitride, and metal selenides, such as zinc selenide, and metal sulfides, such as zinc sulfide, can be used as the anode 103. Anode103 can be modified with plasma-deposited fluorocarbons as disclosed in EP 0914025. For applications where EL emission is viewed only through the cathode electrode, the transmissive characteristics of anode are immaterial and any conductive material can be used, transparent, opaque or reflective. Example conductors for

this application include, but are not limited to, gold, iridium, molybdenum, palladium, and platinum. Typical anode materials, transmissive or otherwise, have a work function of 4.1 eV or greater. Desired anode materials are commonly deposited by any suitable means such as evaporation, sputtering, chemical vapor deposition, or electrochemical means. Anodes can be patterned using well-known photolithographic processes. Optionally, anodes may be polished prior to application of other layers to reduce surface roughness so as to minimize shorts or enhance reflectivity.

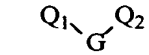
Hole-Injection Layer (HIL):

While not always necessary, it is often useful that a hole-injecting layer 105 be provided between anode 103 and hole-transporting layer 107. The hole-injecting material can serve to improve the film formation property of subsequent organic layers and to facilitate injection of holes into the hole-transporting layer 107. Suitable materials for use in the hole-injecting layer 105 include, but are not limited to, porphyrinic compounds as described in U.S. Patent No. 4,720,432, plasma-deposited fluorocarbon polymers as described in U.S. Patent No. 6,208,075, and some aromatic amines, for example, m-MTDATA (4,4',4"-tris[(3-methylphenyl)phenylamino]triphenylamine). Alternative hole-injecting materials reportedly useful in organic EL devices are described in EP 0 891 121 A1 and EP 1 029 909 A1.

Hole-Transporting Layer (HTL)

The hole-transporting layer 107 of the organic EL device in general contains at least one hole-transporting compound such as an aromatic tertiary amine, where the latter is understood to be a compound containing at least one trivalent nitrogen atom that is bonded only to carbon atoms, at least one of which is a member of an aromatic ring. In one form the aromatic tertiary amine can be an arylamine, such as a monoarylamine, diarylamine, triarylamine, or a polymeric arylamine. Exemplary monomeric triarylamines are illustrated by Klupfel et al. U.S. Patent No. 3,180,730. Other suitable triarylamines substituted with one or more vinyl radicals and/or comprising at least one active hydrogen containing group are disclosed by Brantley et al U.S. Patent Nos. 3,567,450 and 3,658,520.

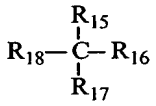
A more preferred class of aromatic tertiary amines are those which include at least two aromatic tertiary amine moieties as described in U.S. Patent Nos. 4,720,432 and 5,061,569. Such compounds include those represented by structural formula (A).



(A)

wherein Q_1 and Q_2 are independently selected aromatic tertiary amine moieties and G is a linking group such as an arylene, cycloalkylene, or alkylene group of a carbon to carbon bond. In one embodiment, at least one of Q_1 or Q_2 contains a polycyclic fused ring structure, e.g., a naphthalene. When G is an aryl group, it is conveniently a phenylene, biphenylene, or naphthalene moiety.

A useful class of triarylamines satisfying structural formula (A) and containing two triarylamine moieties is represented by structural formula (B):

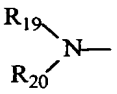


(B)

wherein:

R_{15} and R_{16} each independently represents a hydrogen atom, an aryl group, or an alkyl group or R_{15} and R_{16} together represent the atoms completing a cycloalkyl group; and

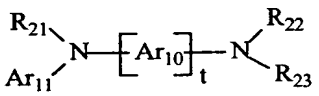
R_{17} and R_{18} each independently represents an aryl group, which is in turn substituted with a diaryl substituted amino group, as indicated by structural formula (C):



(C)

wherein R_{19} and R_{20} are independently selected aryl groups. In one embodiment, at least one of R_{19} or R_{20} contains a polycyclic fused ring structure, e.g., a naphthalene.

Another class of aromatic tertiary amines are the tetraaryldiamines. Desirable tetraaryldiamines include two diarylamino groups, such as indicated by formula (C), linked through an arylene group. Useful tetraaryldiamines include those represented by formula (D):



(D)

wherein

each Ar_{10} is an independently selected arylene group, such as a phenylene or anthracene moiety,

t is an integer of from 1 to 4, and

Ar_{11}, R_{21}, R_{22}, and R_{23} are independently selected aryl groups.

In a typical embodiment, at least one of Ar_4, R_{21}, R_{22}, and R_{23} is a polycyclic fused ring structure, e.g., a naphthalene

The various alkyl, alkylene, aryl, and arylene moieties of the foregoing structural formulae (A), (B), (C), (D), can each in turn be substituted. Typical substituents include alkyl groups, alkoxy groups, aryl groups, aryloxy groups, and halogen such as fluoride, chloride, and bromide. The various alkyl and alkylene moieties typically contain from about 1 to 6 carbon atoms. The cycloalkyl moieties can contain from 3 to about 10 carbon atoms, but typically contain five, six, or seven ring carbon atoms--eg, cyclopentyl, cyclohexyl, and cycloheptyl ring structures. The aryl and arylene moieties are usually phenyl and phenylene moieties.

The hole-transporting layer can be formed of a single or a mixture of aromatic tertiary amine compounds. Specifically, one may employ a triarylamine, such as a triarylamine satisfying the formula (B), in combination with a tetraaryldiamine, such as indicated by formula (D). When a triarylamine is employed in combination with a tetraaryldiamine, the latter is positioned as a layer interposed between the triarylamine and the electron injecting and

transporting layer. Illustrative of useful aromatic tertiary amines are the following:

1,1-Bis(4-di-*p*-tolylaminophenyl)cyclohexane

1,1-Bis(4-di-*p*-tolylaminophenyl)-4-phenylcyclohexane

4,4'-Bis(diphenylamino)quadriphenyl

Bis(4-dimethylamino-2-methylphenyl)-phenylmethane

N,N,N-Tri(*p*-tolyl)amine

4-(di-p-tolylamino)-4'-[4(di-*p*-tolylamino)-styryl]stilbene

N,N,N',N'-Tetra-*p*-tolyl-4-4'-diaminobiphenyl

N,N,N',N'-Tetraphenyl-4,4'-diaminobiphenyl

N,N,N',N'-tetra-1-naphthyl-4,4'-diaminobiphenyl

N,N,N',N'-tetra-2-naphthyl-4,4'-diaminobiphenyl

N-Phenylcarbazole

4,4'-Bis[N-(1-naphthyl)-N-phenylamino]biphenyl

4,4'-Bis[N-(1-naphthyl)-N-(2-naphthyl)amino]biphenyl

4,4"-Bis[N-(1-naphthyl)-N-phenylamino]p-terphenyl

4,4'-Bis[N-(2-naphthyl)-N-phenylamino]biphenyl

4,4'-Bis[N-(3-acenaphthenyl)-N-phenylamino]biphenyl

1,5-Bis[N-(1-naphthyl)-N-phenylamino]naphthalene

4,4'-Bis[N-(9-anthryl)-N-phenylamino]biphenyl

4,4"-Bis[N-(1-anthryl)-N-phenylamino]-*p*-terphenyl

4,4'-Bis[N-(2-phenanthryl)-N-phenylamino]biphenyl

4,4'-Bis[N-(8-fluoranthenyl)-N-phenylamino]biphenyl

4,4'-Bis[N-(2-pyrenyl)-N-phenylamino]biphenyl

4,4'-Bis[N-(2-naphthacenyl)-N-phenylamino]biphenyl

4,4'-Bis[N-(2-perylenyl)-N-phenylamino]biphenyl

4,4'-Bis[N-(1-coronenyl)-N-phenylamino]biphenyl

2,6-Bis(di-*p*-tolylamino)naphthalene

2,6-Bis[di-(1-naphthyl)amino]naphthalene

2,6-Bis[N-(1-naphthyl)-N-(2-naphthyl)amino]naphthalene

N,N,N',N'-Tetra(2-naphthyl)-4,4"-diamino-*p*-terphenyl

4,4'-Bis{N-phenyl-N-[4-(1-naphthyl)-phenyl]amino}biphenyl

4,4'-Bis[N-phenyl-N-(2-pyrenyl)amino]biphenyl

2,6-Bis[N,N-di(2-naphthyl)amine]fluorene

1,5-Bis[N-(1-naphthyl)-N-phenylamino]naphthalene

4,4',4"-tris[(3-methylphenyl)phenylamino]triphenylamine

Another class of useful hole-transporting materials includes polycyclic aromatic compounds as described in EP 1 009 041. Tertiary aromatic amines with more than two amine groups may be used including oligomeric materials. In addition, polymeric hole-transporting/hole injection materials can be used such as poly(N-vinylcarbazole) (PVK), polythiophenes, polypyrrole, polyaniline (Yang, Y. et al. *Appl. Phys. Lett.* 1994, 64, 1245) and copolymers such as poly(3,4-ethylenedioxythiophene) / poly(4-styrenesulfonate) also called PEDOT/PSS(Groenendaal, L. B. et al. *Adv. Mater.* 2000, 12, 481).

Light-Emitting Layer (LEL)

As more fully described in U.S. Patent Nos. 4,769,292 and 5,935,721, the light-emitting layer (LEL) 109 of the organic EL element includes a luminescent or fluorescent material where electroluminescence is produced as a result of electron-hole pair recombination in this region. The light-emitting layer 109 can include a single material including both small molecules and polymers. For small molecules, LEL more commonly consists of a host material doped with a guest compound or compounds where light emission comes primarily from the dopant and can be of any color. The host materials in the light-emitting layer 109 can be an electron-transporting material, as defined below, a hole-transporting material, as defined above, or another material or combination of materials that support hole-electron recombination. The dopant is usually chosen from highly fluorescent dyes, but phosphorescent compounds, e.g., transition metal complexes as described in WO 98/55561, WO 00/18851, WO 00/57676, and WO 00/70655 are also useful. Simultaneously, the color of the EL devices can be tuned using dopants of different emission wavelengths. By using a mixture of dopants, EL color characteristics of the combined spectra of the individual dopant are produced. This dopant scheme has been described in considerable detail for EL devices in U.S. Patent No. 4,769,292 for fluorescent dyes. Dopants are typically coated as 0.01 to 10 % by weight into the host material. Polymeric materials such

as polyfluorenes and poly(arylene vinylenes) (e.g., poly(p-phenylenevinylene), PPV) can also be used as the host material. In this case, small molecule dopants can be molecularly dispersed into the polymeric host, or the dopant could be added by copolymerizing a minor constituent into the host polymer.

An important relationship for choosing a dye as a dopant is a comparison of the bandgap potential which is defined as the energy difference between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of the molecule. For efficient energy transfer from the host to the dopant molecule, a necessary condition is that the band gap of the dopant is smaller than that of the host material. For phosphorescent emitters it is also important that the host triplet energy level of the host be high enough to enable energy transfer from host to dopant.

For small molecules, host and emitting molecules known to be of use include, but are not limited to, those disclosed in U.S. Patent Nos. 4,768,292; 5,141,671; 5,150,006; 5,151,629; 5,405,709; 5,484,922; 5,593,788; 5,645,948; 5,683,823; 5,755,999; 5,928,802; 5,935,720; 5,935,721, and 6,020,078.

For example, small molecule metal complexes of 8-hydroxyquinoline and similar derivatives (Formula E) constitute one class of useful host compounds capable of supporting electroluminescence, and are particularly suitable for light emission of wavelengths longer than 500 nm, e.g., green, yellow, orange, and red.

(E)

wherein:

M represents a metal;

t is an integer of from 1 to 4; and

T independently in each occurrence represents the atoms completing a nucleus having at least two fused aromatic rings.

From the foregoing it is apparent that the metal can be monovalent, divalent, trivalent, or tetravalent metal. The metal can, for example, be an alkali metal, such as lithium, sodium, or potassium; an alkaline earth metal, such as magnesium or calcium; an earth metal, such aluminum or gallium, or a transition metal such as zinc or zirconium. Generally any monovalent, divalent, trivalent, or tetravalent metal known to be a useful chelating metal can be employed.

T completes a heterocyclic nucleus containing at least two fused aromatic rings, at least one of which is an azole or azine ring. Additional rings, including both aliphatic and aromatic rings, can be fused with the two required rings, if required. To avoid adding molecular bulk without improving on function the number of ring atoms is usually maintained at 18 or less.

Illustrative of useful chelated oxinoid compounds are the following:

CO-1: Aluminum trisoxine [alias, tris(8-quinolinolato)aluminum(III)]

CO-2: Magnesium bisoxine [alias, bis(8-quinolinolato)magnesium(II)]

CO-3: Bis[benzo{f}-8-quinolinolato]zinc (II)

CO-4: Bis(2-methyl-8-quinolinolato)aluminum(III)-μ-oxo-bis(2-methyl-8-quinolinolato) aluminum(III)

CO-5: Indium trisoxine [alias, tris(8-quinolinolato)indium]

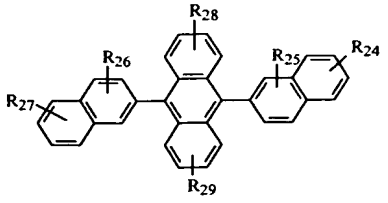
CO-6: Aluminum tris(5-methyloxine) [alias, tris(5-methyl-8-quinolinolato) aluminum(III)]

CO-7: Lithium oxine [alias, (8-quinolinolato)lithium(I)]

CO-8: Gallium oxine [alias, tris(8-quinolinolato)gallium(III)]

CO-9: Zirconium oxine [alias, tetra(8-quinolinolato)zirconium(IV)]

Derivatives of 9,10-di-(2-naphthyl)anthracene (Formula F) constitute one class of useful hosts capable of supporting electroluminescence, and are particularly suitable for light emission of wavelengths longer than 400 nm, e.g., blue, green, yellow, orange or red.



(F)

wherein: R_{24}, R_{25}, R_{26}, R_{27}, R_{28}, and R_{29} represent one or more substituents on each ring where each substituent is individually selected from the following groups:

Group 1: hydrogen, or alkyl of from 1 to 24 carbon atoms;

Group 2: arylof from 5 to 20 carbon atoms;

Group 3: carbon atoms from 4 to 24 necessary to complete a fused aromatic ring of anthracenyl; pyrenyl, or perylenyl;

Group 4: heteroarylof from 5 to 24 carbon atoms as necessary to complete a fused heteroaromatic ring of furyl, thienyl, pyridyl, quinolinyl or other heterocyclic systems;

Group 5: alkoxylamino, alkylamino, or arylamino of from 1 to 24 carbon atoms; and

Group 6: fluorine, chlorine, bromine or cyano.

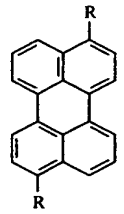
Illustrative examples include 9,10-di-(2-naphthyl)anthracene and 2-*t*-butyl-9,10-di-(2-naphthyl)anthracene. Other anthracene derivatives can be useful as a host in the LEL, including derivatives of 9,10-bis[4-(2,2-diphenylethenyl)phenyl]anthracene.

Distyrylarylene derivatives are also useful hosts, as described in U.S. Patent No. 5,121,029. Carbazole derivatives are particularly useful hosts for phosphorescent emitters.

Polymers incorporating the above small molecule moieties as represented by formulas (E), and (F) are useful host materials. Examples of 9,10-di-(2-naphthyl)anthracene-containing polymers are disclosed U.S. Patent No. 6,361,887.

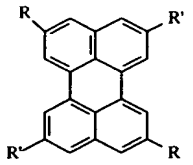
Useful fluorescent dopants (FD) include, but are not limited to, derivatives of anthracene, tetracene, xanthene, perylene, rubrene, coumarin, rhodamine, and quinacridone, dicyanomethylenepyran compounds, thiopyran compounds, polymethine compounds, pyrilium and thiapyrilium compounds, fluorene derivatives, periflanthene derivatives, indenoperylene derivatives, bis(azinyl)amine boron compounds, bis(azinyl)methane compounds, and

carbostyryl compounds. Useful phosphorescent dopants (PD) include but are not limited to organometallic complexes of transition metals of iridium, platinum, palladium, or osmium. Illustrative examples of useful dopants include, but are not limited to, the following:



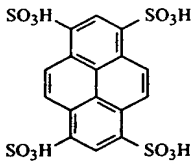
FD 1 R = H

FD 2 R = CO_2Pr-i

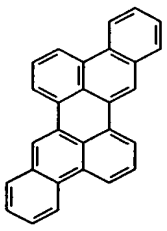


FD 3 R = H, R' = t-Bu

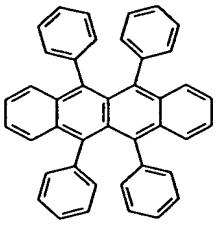
FD 4 R = R' = t-Bu



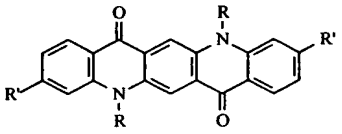
FD 5



FD 6



FD 7



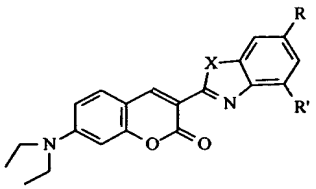
FD 8 R = R' = H

FD 9 R = Me, R' = H

FD 10 R = Pr-i, R' = H

FD 11 R = Me, R' = F

FD 12 R = phenyl, R' =H



FD 13 R = R' = H, X = O

FD 14 R = H, R' = Me, X = O

FD 15 R = Me, R' = H, X = O

FD 16 R = Me, R' = Me, X = O

FD 17 R = H, R' = t-Bu, X = O

FD 18 R = t-Bu, R' = H, X = O

FD 19 R = R' = t-Bu, X = O

FD 20 R = R' = H, X = S

FD 21 R = H, R' = Me, X = S

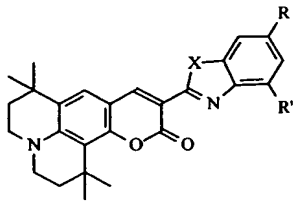
FD 22 R = Me, R' = H, X = S

FD 23 R = Me, R' = Me, X = S

FD 24 R = H, R' = t-Bu, X = S

FD 25 R = t-Bu, R' = H, X = S

FD 26 R = R' = t-Bu, X = S



FD 27 R = R' = H, X = O

FD 28 R = H, R' = Me, X = O

FD 29 R = Me, R' = H, X = O

FD 30 R = Me, R' = Me, X = O

FD 31 R = H, R' = t-Bu, X = O

FD 32 R = t-Bu, R' = H, X = O

FD 33 R = R' = t-Bu, X = O

FD 34 R = R' = H, X = S

FD 35 R = H, R' = Me, X = S

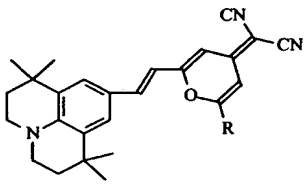
FD 36 R = Me, R' = H, X = S

FD 37 R = Me, R' = Me, X = S

FD 38 R = H, R' = t-Bu, X = S

FD 39 R = t-Bu, R' = H, X = S

FD 40 R = R' = t-Bu, X = S

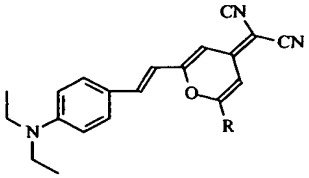


FD 41 R = phenyl

FD 42 R = Me

FD 43 R = t-Bu

FD 44 R = mesityl

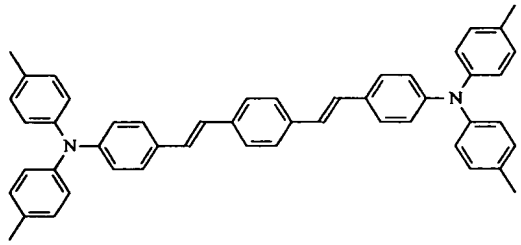


FD 45 R = phenyl

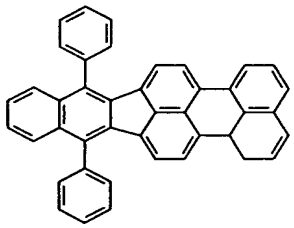
FD 46 R = Me

FD 47 R = t-Bu

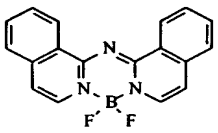
FD 48 R = mesityl



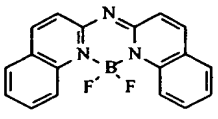
FD 49



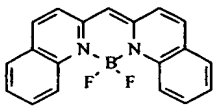
FD 50



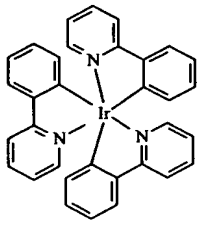
FD 51



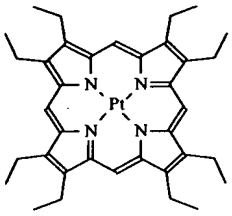
FD 52



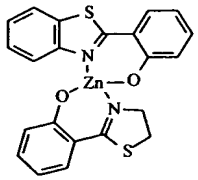
FD 53



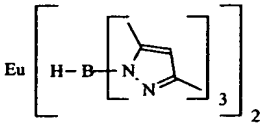
PD 1 ($Ir(PPY)_3$)



PD 2



PD 3



PD 4

Electron-Transporting Layer (ETL):

Preferred thin film-forming materials for use in forming the electron-transporting layer 111 of the organic EL devices of this invention are metal chelated oxinoid compounds, including chelates of oxine itself (also

commonly referred to as 8-quinolinol or 8-hydroxyquinoline). Such compounds help to inject and transport electrons and exhibit both high levels of performance and are readily fabricated in the form of thin films. Exemplary of contemplated oxinoid compounds are those satisfying structural formula (E), previously described.

Other electron-transporting materials include various butadiene derivatives as disclosed in U.S. Patent No. 4,356,429 and various heterocyclic optical brighteners as described in U.S. Patent No. 4,539,507. Triazines are also known to be useful as electron transporting materials. Oxadiazole compounds including small molecules and polymers are useful electron transporting materials as described in U.S. Patent No. 6,451,457.

Cathode

When light emission is viewed solely through the anode, the cathode 113 used in this invention can include nearly any conductive material. Desirable materials have good film-forming properties to ensure good contact with the underlying organic layer, promote electron injection at low voltage, and have good stability. Useful cathode materials often contain a low work function metal (< 4.0 eV) or metal alloy. One preferred cathode material is comprised of a Mg:Ag alloy wherein the percentage of silver is in the range of 1 to 20 %, as described in U.S. Patent No. 4,885,221. Another suitable class of cathode materials includes bilayers comprising a thin electron-injection layer (EIL) in contact with the organic layer (e.g., ETL) which is capped with a thicker layer of a conductive metal. Here, the EIL preferably includes a low work function metal or metal salt, and if so, the thicker capping layer does not need to have a low work function. One such cathode is comprised of a thin layer of LiF followed by a thicker layer of Al as described in U.S. Patent No. 5,677,572. Other useful cathode material sets include, but are not limited to, those disclosed in U.S. Patent Nos. 5,059,861; 5,059,862, and 6,140,763.

When light emission is viewed through the cathode, the cathode must be transparent or nearly transparent. For such applications, metals must be thin or one must use transparent conductive oxides, or a combination of these materials. Optically transparent cathodes have been described in more detail in

U.S. Patent Nos. 4,885,211; 5,247,190, ; 5,703,436; 5,608,287; 5,837,391; 5,677,572; 5,776,622; 5,776,623; 5,714,838; 5,969,474; 5,739,545; 5,981,306; 6,137,223; 6,140,763; 6,172,459; 6,278,236; 6,284,393, JP 3,234,963 and EP 1 076 368,. Cathode materials are typically deposited by evaporation, sputtering, or chemical vapor deposition. When needed, patterning can be achieved through many well known methods including, but not limited to, through-mask deposition, integral shadow masking as described in U.S. Patent No. 5,276,380 and EP 0 732 868, laser ablation, and selective chemical vapor deposition.

Other Useful Organic Layers and Device Architecture

In some instances, layers 109 and 111 can optionally be collapsed into a single layer that serves the function of supporting both light emission and electron transportation or layers 107 and 109 can optionally be collapsed into a single layer that serves the function of supporting both light emission and hole transportation. Alternatively, layers 107, 109 and 111 can optionally be collapsed into a single layer that serves the function of supporting both light emission and hole and electron transportation. This is the preferred EL device structure of this invention and is referred to as "single-layer" device.

It also known in the art that emitting dopants may be added to the hole-transporting layer, which may serve as a host. Multiple dopants may be added to one or more layers in order to create a white-emitting EL device, for example, by combining blue- and yellow-emitting materials, cyan- and red-emitting materials, or red-, green-, and blue-emitting materials. White-emitting devices are described, for example, in EP 1 187 235, EP 1 182 244, U.S. Published Patent Application 20020025419, U.S. Patent Nos. 5,683,823; 5,503,910; 5,405,709, and 5,283,182.

Additional layers such as electron or hole-blocking layers as taught in the art may be employed in devices of this invention. Hole-blocking layers are commonly used to improve efficiency of phosphorescent emitter devices, for example, as in U.S. Published Patent Application 20020015859.

This invention may be used in so-called stacked device architecture, for example, as taught in U.S. Patent Nos. 5,703,436 and 6,337,492.

Deposition of organic layers

The organic materials mentioned above can be deposited as high quality transparent thin films by various methods such as a vapor deposition or sublimation method, an electron-beam method, a sputtering method, a thermal transferring method, a molecular lamination method and a coating method such as solution casting, spin-coating or inkjet printing, with an optional binder to improve film formation. If the material is a polymer, solvent deposition is usually preferred. The material to be deposited by sublimation can be vaporized from a sublimator "boat" often comprised of a tantalum material, e.g., as described in U.S. Patent No. 6,237,529, or can be first coated onto a donor sheet and then sublimed in closer proximity to the substrate. Layers with a mixture of materials can utilize separate sublimator boats or the materials can be pre-mixed and coated from a single boat or donor sheet. Patterned deposition can be achieved using shadow masks, integral shadow masks (U.S. Patent No. 5,294,870), spatially-defined thermal dye transfer from a donor sheet (U.S. Patent Nos. 5,688,551; 5,851,709 and 6,066,357) and inkjet method (U.S. Patent No. 6,066,357).

Preferably, the spin-coating or inkjet printing technique is used to deposit the conjugated polymer of the invention, and only one polymer is deposited in a single layer device.

Encapsulation:

Most organic EL devices are sensitive to moisture or oxygen, or both, so they are commonly sealed in an inert atmosphere such as nitrogen or argon, along with a desiccant such as alumina, bauxite, calcium sulfate, clays, silica gel, zeolites, alkaline metal oxides, alkaline earth metal oxides, sulfates, or metal halides and perchlorates. Methods for encapsulation and desiccation include, but are not limited to, those described in U.S. Patent No. 6,226,890. In addition, barrier layers such as SiOx, Teflon, and alternating inorganic/polymeric layers are known in the art for encapsulation.

Optical Optimization:

Organic EL devices of this invention can employ various well-known optical effects in order to enhance its properties if desired. This includes optimizing layer thicknesses to yield maximum light transmission, providing

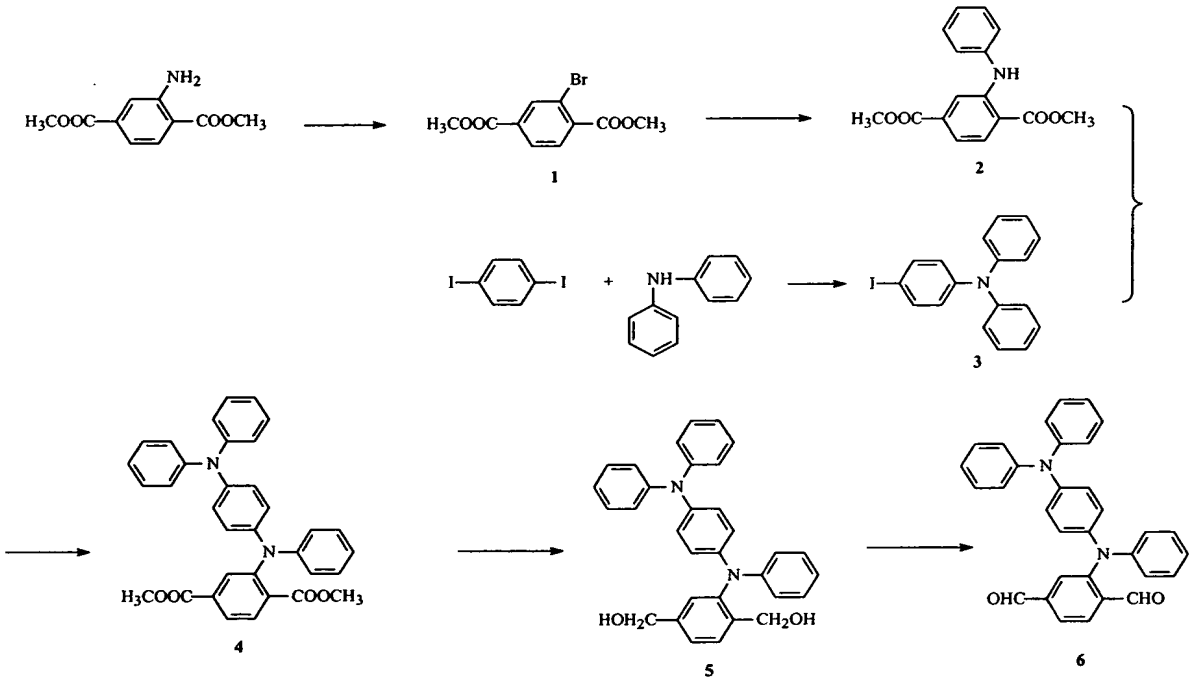
dielectric mirror structures, replacing reflective electrodes with light-absorbing electrodes, providing anti glare or anti-reflection coatings over the display, providing a polarizing medium over the display, or providing colored, neutral density, or color conversion filters over the display. Filters, polarizers, and anti-glare or anti-reflection coatings may be specifically provided over the cover or as part of the cover.

EXAMPLES

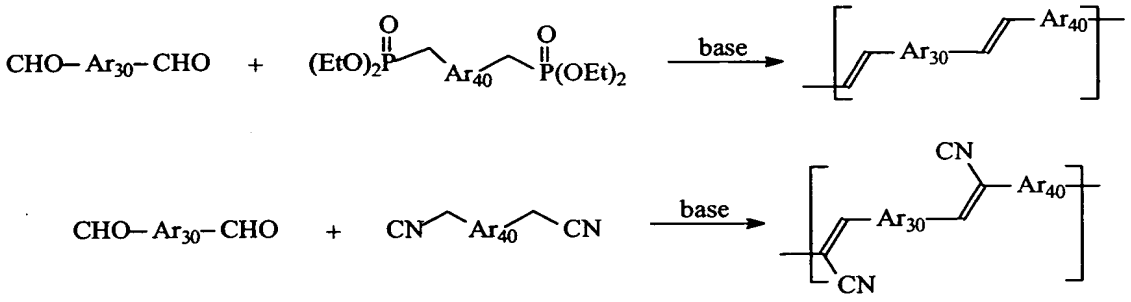
The invention and its advantages are further illustrated by the following specific examples:

Synthesis of Monomers

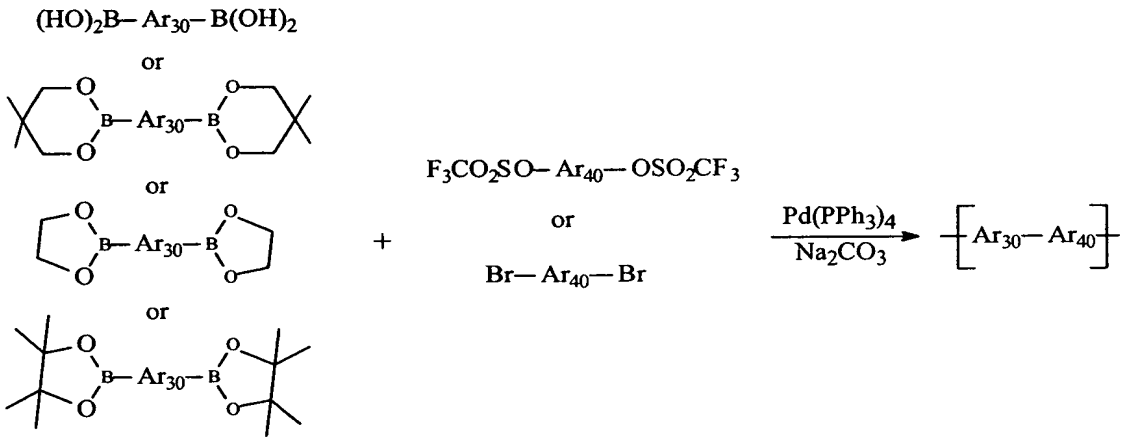
The monomers to be used in the present invention to construct polymers are not necessary to be particularly restricted. Any monomers can be used as long as the polymer formed is a polymer which satisfies the formula (I). Typical synthesis is illustrated in Schemes 1-3.



Scheme 1



Scheme 2



Scheme 3

Example 1: synthesis of dimethyl 2-bromo-terephthalate (compound 1)

Dimethyl 2-amino-terephthalate (10.0 g, 0.048 mol) was dissolved in 60 mL of concentrated HBr solution (50% in water) at 60 °C. The red solution was cooled in an ice-bath and a microcrystalline suspension was obtained. To this suspension was added 2.5 M $NaNO_2$ solution (21 mL, 0.052 mol) under vigorous stirring. The resulting yellow diazonium compound was transferred to a cooled additional funnel (-5 °C) and added to a cooled solution of CuBr (9.1 g, 0.064 mol) in 25 mL of concentrated HBr solution. A defoaming agent n-butanol was used to prevent excessive foaming. After the addition was complete, the reaction was heated to 70 °C until no further nitrogen evolved. The reaction was cooled, and was extracted with ether. The organic phase was washed with water and dried

over $MgSO_4$. The crude product was obtained as gray black solid and was purified by recryllization from heptane to give 6.6 g of pure product as white solid at 51% yield. ^1H NMR ($CDCl_3$) δ (ppm): 3.95 (s, 6 H), 7.82 (d, J = 8.1 Hz, 1 H), 8.01 (dd, J_1 = 8.1 Hz, J_2 = 1.5 Hz, 1 H), 8.32 (d, J = 1.5 Hz, 1 H). ^{13}C NMR ($CDCl_3$) δ (ppm): 52.64, 52.71, 121.36, 128.01, 130.94, 133.60, 135.09, 135.99, 164.89, 166.02. Mp 48-50 °C. FD-MS: *m/z* 273 (M^+).

Example 2: synthesis of dimethyl 2-phenylamino-terephthalate (compound 2)

Dimethyl 2-bromo-terephthalate (10.0 g, 0.037 mol), aniline (17.0 g, 0.18 mol), potassium phosphate (11.7 g, 0.055 mol), and $Pd_2(dba)_3$ (0.67 g, 0.73 mmol) were mixed in 100 mL of anhydrous toluene. The mixture was bubbled with nitrogen for 10 min. and tri *t*-butyl phosphine (0.12 g, 0.58 mmol) was added. The reaction was heated to reflux overnight. The reaction was cooled down and extracted with ether. The combined organic phase was dried over $MgSO_4$ and solvent was removed. The crude product was obtained as dark brown oil. The crude product was purified by column on silica gel using 10/90 methylene chloride/heptane as an eluent to obtain yellow solid which was further recrystallized in heptane to give 6.0 g of pure product as orange crystals at 58% yield. ^1H NMR ($CDCl_3$) δ (ppm): 3.87 (s, 3 H), 3.93 (s, 3 H), 7.24-7.36 (m, 6 H), 7.91-8.03 (m, 2 H), 9.50 (s, br, 1H). ^{13}C NMR ($CDCl_3$) δ (ppm): 52.06, 52.34, 114.83, 115.16, 117.24, 122.51, 124.01, 129.54, 131.75, 134.85, 140.16, 147.74, 166.54, 168.36. Mp 85-87 °C. FD-MS: *m/z* 285 (M^+).

Example 3: synthesis of 4-diphenylamino-iodobenzene (compound 3)

Diphenyl amine (21.0g, 0.12 mol), 1,4-diiodobenzene (49.1 g, 0.15 mol), potassium carbonate (51.4 g, 0.37 mol), copper bronze (15.6 g, 0.25 mol), and crown-18-6 (3.1 g, 15 wt% to diphenyl amine) were mixed in 200 mL of o-dichlorobenzene and the reaction was heated to reflux overnight. The reaction was cooled down and the solid was filtered off and washed with methylene chloride. The filtrate was concentrated and cooled in dry ice. 1,4-Diiodobenzene crashed out upon cooling and was filtered off. The process was repeated until most of 1,4-diiodobenzene was removed from crude product. The crude product was then purified by column chromatography on silica gel using heptane as an

eluent to give 20.1 g of pure product as white solid at 44% yield. ^{1}H NMR ($CDCl_3$) δ (ppm): 6.82 (d, J=8.8 Hz, 2 H), 7.00-7.27 (m, 10 H), 7.48 (d, J = 8.8 Hz). ^{13}C NMR ($CDCl_3$) δ (ppm): 122.67, 123.30, 124.13, 124.52, 125.27, 129.17, 129.34, 138.01, 147.22, 147.69, 147.82. Mp 102-104 °C. FD-MS: *m/z* 371 (M^+).

Example 4: synthesis of dimethyl 2-(4-diphenylaminophenyl)phenylamino-terephthalate (compound 4)

Compound **2** (5.0 g, 0.018 mol), compound **3** (7.8 g, 0.021 mol), potassium carbonate (7.3 g, 0.052 mol), cupper bronze (2.2 g, 0.035 mol) and crown-18-6 (0.75 g) were mixed in 50 mL of o-dichlorobenzene and heated to reflux overnight. After cooling down, the solid was filtered off and the reaction was extracted with ether. The combined organic phase was dried over $MgSO_4$. The crude product was obtained as dark brown oil and was purified by column chromatography on silica gel using 25/75 methylene chloride/heptane to give 4.6 g of pure product as orange foam at 55% yield. ^{1}H NMR ($CDCl_3$) δ (ppm): 3.58 (s, 3 H), 3.95 (s, 3 H), 7.00-7.10 (m, 9 H), 7.18 (d, J = 7.6 Hz, 4 H), 7.27-7.34 (m, 6H), 7.76 (d, J =8.0 Hz, 1 H), 7.62 (dd, J_1 = 8.0 Hz, J_2 = 1.4 Hz, 1 H), 7.98 (d, J =8.0 Hz, 1 H). ^{13}C NMR ($CDCl_3$) δ (ppm): 51.88, 52.27, 122.22, 122.26, 122.34, 123.60, 124.467, 125.26, 129.06, 129.56, 131.06, 132.46, 133.89, 142.34, 142.88, 146.60, 147.36, 147.63, 165.82, 167.16; Mp 139-140 °C. FD-MS: *m/z* 528 (M^+).

Example 5: synthesis of 2-(4-diphenylaminophenyl)phenylamino-1,4-dihydroxymethylbenzene (compound 5)

Compound **4** (12.3 g, 0.023 mol) was dissolved in 100 mL of dry THF and added slowly to a cooled $LiAlH_4$ (1.9 g, 0.051 mol) in 100 mL of dry THF suspension. After the addition, the reaction was heated to reflux for 1 h. The reaction was cooled down and quenched with $Na_2SO_4.10H_2O$. The reaction was then filtered to give 10.4 g of pure product as off-white solid at 95% yield. ^{1}H NMR ($CDCl_3$) δ (ppm): 4.37 (s, 2 H), 4.56 (s, 2 H), 6.79-6.92 (m, 9 H), 6.99 (d, J = 8.2 Hz, 4 H), 7.09-7.18 (m, 8 H), 7.42 (d, J = 7.8 Hz, 1 H). ^{13}C NMR ($CDCl_3$) δ (ppm): 62.09, 64.65, 121.12, 121.60, 122.37, 123.27, 123.62, 124.58, 125.49, 127.50, 129.16, 129.20, 129.58, 137.49, 142.02, 142.33, 142.45, 144.57, 147.64, 147.75; Mp 158-160 °C. FD-MS: *m/z* 472 (M^+).

Example 6: synthesis of 2-(4-diphenylaminophenyl)phenylamino-1,4-diformyllbenzene (compound 6)

Compound **5** (0.89 g, 1.9 mmol) was dissolved in 15 mL of methylene chloride and pyridinium chlorochromate (PCC, 0.89 g, 4.1 mmol) was added. The reaction was stirred at room temperature for 3 h and quenched with water. The reaction was filtered through a pad of Celite and washed with methylene chloride. The filtrate was separated and the aqueous layer was extracted with methylene chloride and the organic phase was dried over $MgSO_4$. The crude product was obtained as black solid and was purified by column on silica gel using 30/70 ether/heptane as an eluent to give dark red foaming solid. The pure product was obtained after further recrystallization from ethanol to give 0.25 g of bright orange crystals at 17% yield. ^1H NMR ($CDCl_3$) δ (ppm): 6.83-7.03 (m, 14 H), 7.16-7.21 (m, 5 H), 7.59-7.62 (m, 2 H), 7.90 (d, J = 7.8 Hz, 1 H). ^{13}C NMR ($CDCl_3$) δ (ppm): 122.56, 122.88, 123.27, 124.17, 124.76, 124.87, 124.99, 129.02, 129.27, 129.75, 130.12, 135.02, 141.05, 142.81, 143.92, 147.48, 148.59, 150.96, 189.96, 191.23. Mp 159-161 °C. FD-MS: *m/z* 468 (M^+).

Synthesis of Polymers

Example 7: General procedure for a Horner-Emmons reaction:

Equimolar of dicarboxyaldehyde and diphosphate monomers were dissolved in anhydrous THF under nitrogen. To this solution was added 2.5 equivalent of NaH. The reaction was stirred at room temperature overnight under nitrogen. Small amount of benzaldehyde was added to endcap phosphate endgroup. The polymer was precipitated into methanol, filtered, re-dissolved in chloroform and precipitated twice more from methanol. The resulting polymer was dried under vacuum at 45 °C overnight.

Example 8: General procedure for a Knoevenagel reaction:

Equimolar of dicarboxyaldehyde and dicyano monomers were dissolved in a mixed solvent of 1:1 anhydrous THF and t-butyl alcohol under nitrogen. To this solution was added catalytic amount of potassium t-butoxide. The reaction was stirred at room temperature overnight under nitrogen. The polymer was precipitated into methanol, filtered, re-dissolved in chloroform and

precipitated twice more from methanol. The resulting polymer was dried under vacuum at 45 °C overnight.

EL Device Fabrication and Performance

Example 9

An EL device satisfying the requirements of the invention was constructed in the following manner. The organic EL medium has a single layer of the organic compound described in this invention.

a) An indium-tin-oxide (ITO) coated glass substrate was sequentially ultra-sonicated in a commercial detergent, rinsed with deionized water, degreased in toluene vapor and exposed to ultraviolet light and ozone for a few minutes.

b) An aqueous solution of PEDOT (1.3% in water, Baytron P Trial Product AI 4083 from H. C. Stark) was spin-coated onto ITO under a controlled spinning speed to obtain thickness of 500 Angstroms. The coating was baked in an oven at 110 °C for 10 min.

c) A toluene solution of a polymer (300 mg in 30 mL of solvent) was filtered through a 0.2 μm Teflon filter. The solution was then spin-coated onto PEDOT under a controlled spinning speed. The thickness of the film was between 500-1000 Angstroms. On the top of the organic thin film was deposited a cathode layer consisting of 15 angstroms of a CsF salt, followed by a 2000 angstroms of a 10:1 atomic ratio of Mg and Ag.

The above sequence completed the deposition of the EL device. The device was then hermetically packaged in a dry glove box for protection against ambient environment.

Table 1 summarizes the characterization of the polymers prepared in the present invention. Absorption (AB) and photoluminescence (PL) spectra were obtained from solid thin films of the polymers and EL spectra were obtained from ITO/PEDOT/polymer/CsF/Mg:Ag EL devices. The fabrication of EL devices was illustrated in Example 9. FIG. 2 shows EL spectra of polymer 5, 28, and 58. FIG. 3 shows AB and PL spectra of polymer 5 in dilute toluene solution

and thin film. FIG. 4 And FIG. 4 shows the voltage-current-luminance characteristics of the EL device of polymer 5.

Table 1. Characterization of polymers according to Examples.

Polymer	M_w [a]	PDI	T_d (°C)	T_g (°C)	AB [b] (λ_{max} nm)	PL [c] (λ_{max} nm)	EL (λ_{max} nm)
5	12200	1.83	417	108	319, 463	564 (460)	552
28	24700	3.18	362	107	314	381 (310)	584
58	77900	7.64	399	152	309	367 (310)	556

[a] weight average molecular weight, determined by size exclusion chromatography in THF using polystyrene standard. [b] as solid state thin film [c] as solid state thin film, the number in the parenthesis is the excitation wavelength.

It will be understood that organic layers in accordance with the invention can be an emissive layer or a hole injection layer or both.

The invention has been described in detail with particular reference to certain preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit and scope of the invention.

PARTS LIST

101 Substrate

103 Anode

105 Hole-Injecting layer (HIL)

107 Hole-Transporting layer (HTL)

109 Light-Emitting layer (LEL)

111 Electron-Transporting layer (ETL)

113 Cathode

250 Current / Voltage source

260 Electrical conductors